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BRIEF

DDT Residues in Starlings, 1974

Paul R. Nickerson [1] and Kyle R. Barbehenn [2]

ABSTRACT

In the preceding issue of this journal, the authors suggested that the mean level of DDT plus metabolites in starlings should drop below 0.1 ppm for the 1974 collection. They based their prediction on an analysis of the relationship between mean levels of DDT and its metabolites in starlings and estimates of domestic disappearance of DDT. The present brief summarizes initial findings from the 1974 starling collection. Authors indicate that their earlier estimates for disappearance of total DDT were optimistic: the geometric mean for 1974 was 0.282, a 36 percent reduction from the 1972 mean of 0.442.

Introduction

Based upon an analysis of the relationship between mean levels of DDT and its metabolites in starlings and estimates of domestic disappearance of DDT, authors suggested in the preceding issue of this periodical that the residue levels of these organochlorines in starlings should drop below a mean of 0.1 ppm for the 1974 collection (*1*).

Analytical Results

Residue analysis results from the 1974 collection are now in hand and it is apparent that the extrapolation from a small data base was overly optimistic. The geometric mean of DDT plus metabolites is 0.282 ppm for 1974.

Of the 122 sites sampled, 17 of the values exceeded 1.0 ppm and 2 (3G3 in Arkansas and 4C1 in Arizona; see Tables 1,3) reached a level of 9.2 ppm. Although the reduction of 0.160 ppm (36 percent) from the mean level of 1972 (0.442 ppm) is substantial it is clear that DDT remains by far the most abundant source of pesticide residues found in starlings two full growing seasons after the major uses of DDT were cancelled.

LITERATURE CITED

(*1*) *Nickerson, Paul R., and Kyle R. Barbehenn. 1975.* Organochlorine residues in starlings, 1972. Pestic. Monit. J. 8(4):247-254.

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RESIDUES IN FISH, WILDLIFE, AND ESTUARIES

Chlorinated Pesticides and Polychorinated Biphenyls in Marine Species, Oregon/Washington Coast, 1972 [1]

Robert R. Claeys,[2] Richard S. Caldwell,[3] Norman H. Cutshall,[4] and Robert Holton[4]

ABSTRACT

Concentrations of chlorinated pesticides and polychlorinated biphenyls (PCB's) were determined in three offshore marine species from the Oregon/Washington coast: pink shrimp, euphausiids, and flatfish; five species of bivalve mollusks from five estuaries along the Oregon coast; several fish species from the Coos Bay and Columbia River estuaries; and a summer run of steelhead from the Rogue River.

The compounds p,p'-DDE and PCB's were detected most frequently. Euphausiids and pink shrimp contained approximately 2 ppb (μg/kg) wet-weight DDE and 8 and 25 ppb PCB's, respectively. Offshore flatfish contained an average of 9 ppb DDE and 29 ppb PCB's. DDE residues in estuarine mollusks approximated 0.5 ppb. PCB levels were not detectable (<3 ppb) except in collections from the mouth of the Columbia River where levels averaged 400 ppb PCB's and 17 ppb DDT. Selected Columbia River fish species contained 38 ppb DDE and 480 ppb PCB's; summer-run steelhead in the Rogue River contained 97 ppb DDE and 125 ppb PCB's.

PCB chromatograms of most euphausiids closely resembled those of Aroclor 1254. Chromatograms of shrimp and flatfish indicated selective metabolism of two compounds in the Aroclor 1254 formulation. Biphenyls of higher chlorine content were also detected in the shrimp and flatfish.

Introduction

A global program to determine baseline levels of metals, hydrocarbons, and chlorinated hydrocarbons was initiated in 1971 by the International Decade for Ocean Exploration (IDOE) Program of the National Science Foundation. Baseline data for chlorinated hydrocarbons in the North Pacific Ocean are reported here. In addition, baseline levels in mollusks were determined in several Oregon estuaries as part of the National Estuarine Monitoring Program. Several species of fish were collected from two of these estuaries along with some summer-run steelhead (*Salmo gairdnerii*), a type of rainbow trout, from the Rogue River. Chlorinated hydrocarbon levels obtained under the IDOE Program from the Atlantic Ocean (*1*) and the Gulf of Mexico (*2*) surveys have already been published.

Sampling and Analytical Procedures

Pink shrimp (*Pandalus jordani*), euphausiids (*Euphausia pacifica*), and several species of flatfish were collected at ocean stations from Newport, Oreg., to the Straits of Juan de Fuca during September and October 1971 (Fig. 1). An otter trawl and an Isaacs-Kidd midwater trawl were used in these collections.

Estuarine bivalves were collected quarterly from December 1971 through October 1972 in five Oregon estuaries: Columbia River, Tillamook Bay, Yaquina Bay, Umpqua River estuary, and Coos Bay (Fig. 1). Species collected were the cockle clam (*Clinocardium nuttallii*), Eastern softshell clam (*Mya arenaria*), bay mussel (*Mytilus edulis*), Asiatic clam (*Corbicula fluminea*), and a species of Anodonta. The latter two species inhabit only fresh water and were the most abundant mollusks in the Columbia River; estuarine clams were not readily available. In addition, several species of estuarine fish were collected in the Coos Bay and Columbia River estuaries during January 1973 and

[1] Research conducted under National Science Foundation International Decade of Ocean Exploration Grant 6X28744, National Marine Fisheries Contract N-042-14-72(N), and U.S. DHEW Public Health Service Grant ES00040.

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August 1972, respectively; summer-run steelhead were obtained from the Rogue River in September 1970. Except for the steelhead, samples were frozen in glass jars washed with acetone. Special care was taken to avoid contamination because of the possible presence of polychlorinated biphenyls (PCB's) aboard ship.

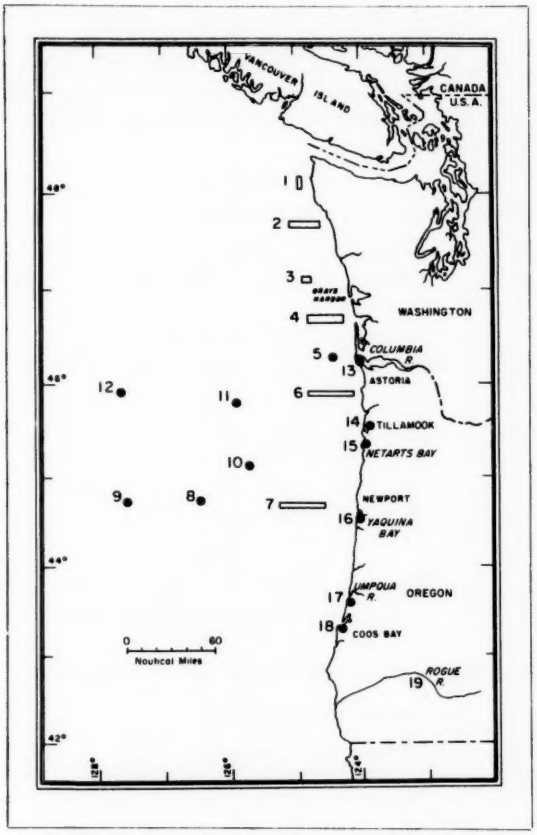

FIGURE 1. *Stations on Oregon/Washington coast sampled for residues in marine species*

Analytical procedures were similar to those of Porter et al. (*3*) except for hexane-acetonitrile partitioning; for that, analysts followed the method of Giuffrida et al. (*4*). Briefly, the shrimp, euphausiids, and small fish were ground whole in a meat grinder and a subsample not exceeding 3 g lipid or 100 g tissue was taken for analysis. Steelhead were analyzed individually by taking a cross section posterior to the anal opening. Mollusks were prepared and extracted as described by Butler (*5*). They were ground with a desiccant mixture of 10 percent QUSO (precipitated silica) and 90 percent anhydrous sodium sulfate. The sample and desiccant were mixed at an exact ratio of 1:3 by weight before taking a 120-g subsample. Fish and shrimp were extracted in a blendor with 2:1 hexane:acetone (v/v) and mollusks were extracted by Soxhlet with 1:1 hexane:acetone. A 25- 50-g sample was extracted, the solvent evaporated, and the residue weighed for an approximate lipid content.

Approximately 90 percent of the lipid was separated from the organochlorines by chromatographic column elution (*4*). The extract was evaporated under a stream of air and the lipid residue was mixed with florisil and loaded on a dry-packed florisil column. Pesticides were then eluted with 9:1 acetonitrile:water and partitioned into hexane after aqueous dilution. Additional cleanup was obtained on a second florisil column. PCB's, DDT, BHC, chlordane, mirex, and toxaphene compounds were eluted from the second column with 5 percent benzene in hexane (v/v). Dieldrin, endrin, heptachlor epoxide, and methoxychlor were eluted with hexane containing 10 percent ethyl ether and 0.25 percent acetone (v/v).

Major PCB isomers were separated from pesticides by a modification of the procedure of Armour and Burke (*6*) by substituting a 1 percent water deactivated silicic acid column. The 4-10 chloro PCB compounds were eluted with hexane and the 1-3 chloro PCB's and pesticide compounds were eluted with 5 percent aqueous methanol. Pesticides were partitioned into hexane after aqueous dilution of the methanol.

Compounds were normally separated and quantitated by gas-liquid chromatography on 122-cm-by-3-mm-ID pyrex columns filled with a 2:1 mixture of 7 percent QF-1 and 7 percent DC-11 liquid phases on high-performance chromosorb W, 100/120 mesh, or with 7 percent DC-11. Columns were operated at 195° C with 20 ml/min N_2 flow. The flow rate employed was 1.5 times the optimum rate for maximum PCB resolution. Both electron-capture and microcoulometric-halide detectors were employed. Sensitivity of the microcoulometric detector was 1-3 ng dieldrin or DDE.

Base hydrolysis was used to confirm the presence of DDD and DDT in selected samples by conversion to DDE and DDMU, respectively (*7*). PCB's remain stable although the α and γ BHC isomers are destroyed during this procedure. A 1:1 fuming HNO_3:concentrated sulfuric acid nitration test (*8*) was used to confirm the presence of chlordane and toxaphene; these are the only compounds which are not nitrated.

Special precautions were employed to improve analysis of low pesticide concentrations. Glassware was baked at 250°-300° C in a large oven (*9*) and other items such as glass wool, sodium sulfate, and florisil were baked at 450° C in a muffle furnace to reduce blank levels. In addition, blanks were analyzed before any samples were begun.

For PCB quantitation peak heights of the sample and standard were added. When peaks were missing, a zero was included in the summation. For two different reasons PCB values may be low: no attempt was made to

identify 8-10 chloro biphenyls; and 1-3 chloro biphenyls elute with the pesticide fraction from the silicic acid column.

Selected samples were spiked with known standards prior to extraction at a concentration of 10 higher than that previously analyzed. Mean recoveries were 75 percent for DDE, 63 percent for DDT, 83 percent for dieldrin, and 100 percent for Aroclor 1260 when silicic acid column separation of PCB's was employed (*10*). Recoveries from the silicic acid column were only 85 percent for DDE and DDT, thus accounting for low DDT recovery. Mean recovery of DDE for mollusks was 92 percent without silicic acid column separation. Values reported here are uncorrected for recovery or blank levels, except for mollusks, in which case blank levels were subtracted.

Results

Levels of chlorinated hydrocarbons in offshore species, estuarine fish, and Rogue River steelhead are given in Tables 1 and 2; offshore results are summarized in Table 3. Most of these data were presented in 1972 at a workshop of the International Decade of Ocean Exploration (*10*).

TABLE 1. *Chlorinated hydrocarbon concentrations in marine species, Washington/Oregon—1972*

Species		Concentration, µg/kg wet weight				
		p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	Total DDT	PCB's
Euphausia pacifica	Samples analyzed	14	14	14	14	11
	Samples with residues	14	5	5	14	11
	Mean	2.2	0.2	0.6	3.0	7.5
	Range	0.2-5.8	0-0.6	0-4.0	0.2-5.9	1-22
Pandalus jordani	Samples analyzed	13	13	13	13	13
	Samples with residues	13	8	8	13	13
	Mean	1.9	0.3	0.5	2.7	25
	Range	0.9-3.7	0.2-1.0	0.2-3.0	1.1-5.0	11-69
Flatfish (genus and species unknown)	Samples analyzed	13	13	13	13	10
	Samples with residues	13	12	12	13	10
	Mean	8.5	1.0	1.0	10.5	29
	Range	3.4-18	0.7-1.7	0.6-2.0	4-19.7	16-121
Blanks	Samples analyzed	11	11	11	9	9
	Samples with residues	11	5	4	7	7
	Mean	0.06	0.03	0.03	0.12	<2
	Range	0.02-0.13	0-0.12	0-0.10	0-0.25	0-4

TABLE 2. *Chlorinated hydrocarbons in selected marine species collected off Oregon/Washington coast, September 1971*

Sampling Station	Location: Latitude, Longitude	Concentration, µg/kg wet weight						
		p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	Total DDT	Dieldrin	Aroclor 1254	Aroclor 1260
		Euphausia pacifica						
7	44° 39′ 124° 31′	1.0	0.6	0.9	3		5	
7	44° 43′ 124° 41′	5.8			6		5	
7	44° 39′ 124° 52′	4.5			5		6	
7	44° 41′ 125° 09′	1.1			1		8	3
7	44° 39′ 125° 14′	3.3			3			3
8	44° 43′ 126° 29′	2.6			3		22	
8	44° 43′ 126° 29′	0.8			1		8	
9	44° 43′ 127° 35′	1.0			1		25	5
10	45° 10′ 125° 39′	1.1			1		4	
11	45° 46′ 125° 49′	2.3	0.1	0.5	3			13
12	45° 56′ 127° 40′	3.3	0.4[1]	0.5[1]	4			1
5	46° 21′ 124° 27′	1.8			2		4	
4	46° 31′ 124° 30′	0.2[2]			0.2		NA	

(Continued next page)

TABLE 2 (cont'd.). *Chlorinated hydrocarbons in selected marine species collected off Oregon/Washington coast, September 1971*

Sampling Station	Location: Latitude, Longitude	Concentration, µg/kg wet weight						
		p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	Total DDT	Dieldrin	Aroclor 1254	Aroclor 1260
4	46° 41′ 124° 46′	2.1 [2]	0.6 [2]	3 [2]	7	5 [2]	NA	
3	47° 11′ 124° 48′	1.3 [1]	0.6 [1]	4 [1]	6	4 [1]	NA	
		Pandalus jordani						
7	44° 39′ 124° 35′	2.4	0.7	0.3	3		9	10
7	44° 43′ 124° 41′	3.7	0.2	0.3	4		11	
6	45° 55′ 124° 14′	1.1			1		12	
6	45° 55′ 124° 28′	1.2			1		36	8
6	45° 56′ 124° 41′	2.0			2		9	
5	46° 21′ 124° 27′	0.9	0.5	0.3	2		35	10
4	46° 37′ 124° 26′	2.5	0.3	0.2	3		33	
4	46° 39′ 124° 39′	1.4			1		14	
4	46° 41′ 124° 37′	1.8	0.4	0.6	3		16	
3	47° 06′ 124° 41′	1.9	1.0		3		19	50
3	47° 06′ 124° 47′	1.0	0.5	0.2	2		15	
3	47° 11′ 124° 48′	2.0 [2]		3 [2]	5		44	
2	47° 39′ 125° 05′	3.0	0.3 [1]	1.1 [1]	4		20	
		Sand Shrimp						
6	45° 55′ 124° 14′	<1.3			1		ND	
2	47° 39′ 124° 36′	<0.3					7	
2	47° 40′ 124° 54′	1.1	0.3	0.3	2		28	
1	48° 06′ 124° 51′						6	
		Sergestid Shrimp						
7	44° 43′ 124° 41′	25		4.1	29		17	
		Galathea Shrimp						
4	46° 41′ 124° 37′	1.4			5		5	
		Flatfish						
6	45° 54′ 124° 02′	6.4	0.7	1.1	8		ND	12
6	45° 55′ 124° 14′	10.4	1.0	1.1	13		52	
6	45° 56′ 124° 12′	10 [2]	<3 [2]	<5 [2]	18		NA	
6	45° 55′ 124° 28′	12.6	0.9	0.8	14		24	
6	45° 56′ 124° 41′	12.7	1.7 [1]	1.8 [1]	25		28	
4	46° 37′ 124° 14′	11.0	1.8 [1]	2.0 [1]	15		85	36
4	46° 37′ 124° 26′	3.4	1.0	1.2	6		25	
2	47° 28′ 124° 41′	6.7	1.0	1.5	9		28	
2	47° 39′ 124° 36′	5.0	0.9		6		24	

(Continued next page)

TABLE 2 (cont'd.). *Chlorinated hydrocarbons in selected marine species collected off Oregon/Washington coast, September 1971*

Sampling Station	Location: Latitude, Longitude	Concentration, μg/kg wet weight						
		p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	Total DDT	Dieldrin	Aroclor 1254	Aroclor 1260
2	47° 39′ 124° 36′	5.7	NA	NA	6		12	
2	47° 40′ 124° 54′	10.0	0.6	0.7	17		18	
2	47° 41′ 124° 46′	5.7	0.9	0.7	7		16	
2	47° 39′ 125° 05′	17.4	0.8	0.9	19		30	
2	47° 39′ 125° 05′	16.6	0.8	0.9	18		18	
2	47° 39′ 125° 05′	20.0	NA	NA	20		21	
1	48° 06′ 124° 51′	4.3	0.8	0.9	6		16	
		Salpa						
9	44° 43′ 127° 35′	0.1					20	
		Clupea harengus pallasii						
4	46° 37′ 124° 14′	19	4.0	1.6	25		146	
		Small Fish						
1	48° 12′ 124° 56′	107	2 [1,2]	6 [1,2]	115	3 [1,2]	NA	

NOTE: NA = not analyzed.
ND = no data because of interference with analytical process.
Blank spaces imply residues below detectable levels.
Representative *Euphausia pacifica* sample contained 1.8 percent lipid; *Pandalus jordani*, 2.0 percent; sand shrimp, 2.4 percent.
[1] Alcoholic base hydrolysis.
[2] Microcoulometric detector confirmation.

TABLE 3. *Chlorinated hydrocarbons in estuarine fish and Rogue River steelhead, 1970-73*

Species	Concentration, μg/kg wet weight						
	p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	Total DDT	Dieldrin	Aroclor 1254	Aroclor 1260
Coos Bay Estuary, January 1973, Station 18							
Striped Seaperch *(Embiotoca lateralis)*	5			5		23 [1]	
Sand Sole *(Psettichthys melanostictus)*	7			7		26 [1]	
Staghorn Sculpin *(Leptocottus armatus)*	3			3		14 [1]	
Starry Flounder *(Platichthys stellatus)*	6			6			27 [1]
Blank	0.02			0.02	NA	2	
Percent recovery of sample spiked at 17 ppb			87		81		
Columbia River Estuary, August 1972, Station 13 [2]							
Starry Flounder *(Platichthys stellatus)*	18	8	8	34	310		
Tom Cod *(Microgadus proximus)*					90		
Peamouth Chub *(Mylochilus causinus)*	81	62		143	1160		
Finescale Sucker *(Catostomus syncheilus)*	14	28	11	53	350		
Blank	<1	<1	<1		<1		
South Rogue River, September 1970, Station 19							
Steelhead *(Salmo gairdnerii)* [3]							
2 [4]	73			73	28	100	
9	110			110	15	150	
1,6 [5]	140			140	9	NA	
8,10 [5]	62			62	29	NA	
5,7 [5]	72			72		NA	
3,4 [5]	126			126	24	NA	

NOTE: NA = not analyzed.
Representative starry flounder sample contained 2.5 percent lipid; representative steelhead contained 10.6 percent lipid.
Blank spaces imply residues below detectable levels.
[1] Microcoulometric confirmation.
[2] Aroclor 1254 concentrations not reported for species from station 13 because of interference with analytical process.
[3] South Rogue River column 1 shows Oregon State University identification numbers for samples not identified by species.
[4] Species also contained 6 μg/kg chlordane and 6 μg/kg thiodan.
[5] Sample too small for PCB analysis.

Offshore collections showed little geographical differences. DDE was frequently the only DDT-related compound present, averaging about 2 ppb in euphausiids and pink shrimp and 9 ppb in flatfish. PCB levels were slightly higher, averaging 8, 25, and 29 ppb for euphausiids, pink shrimp, and flatfish, respectively.

Figure 2 compares a typical shrimp chromatogram with those of Aroclor 1254 and 1260. Although most euphausiid PCB chromatograms resembled Aroclor 1254, peaks 21 and 23 were low or absent in pink shrimp and flatfish. Peak number 34 was also absent but peak 37 was usually present. Tentative indentification has been previously reported (*11*). Peaks 21 and 23 are both five chlorobiphenyls; peaks 34 and 37 are seven chlorobiphenyls.

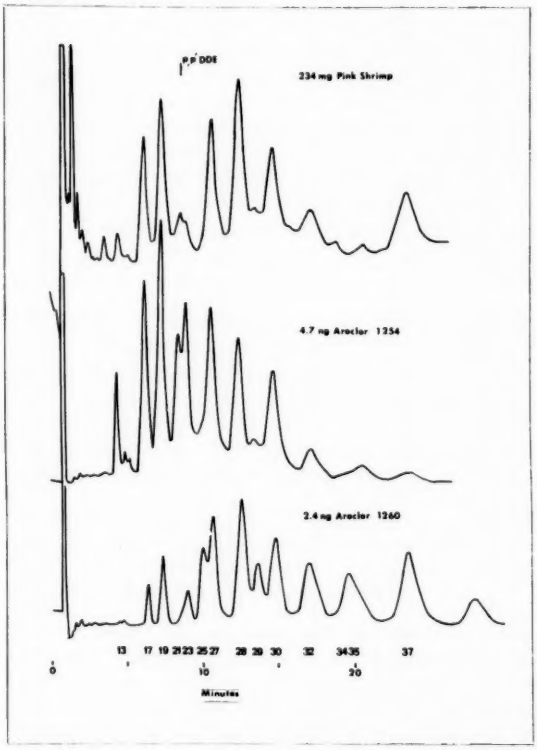

FIGURE 2. *Typical chromatograms of the PCB fraction, pink shrimp extract*

Dieldrin may have been present in the offshore species; but because of poor lipid separation, many peaks were present in the eluant containing dieldrin, making positive identification and quantitation difficult. In about one-half the pink shrimp and euphausiid collections and nearly all flatfish samples an apparent dieldrin peak was present. A second chromatographic column, 3 percent diethylene glycol succinate, was used for further confirmation.

Where dieldrin was indicated, levels were 0.2-0.5 ppb except for two euphausiid samples which contained about 5 ppb. In these latter two samples microcoulometric chloride detection positively confirmed dieldrin. These samples, collected 25 miles west of Grays Harbor (sampling stations 3 and 4), also contained higher levels of DDT and DDD. Dieldrin was also positively confirmed in a small unidentified fish collected near the Straits of Juan de Fuca (station 1). Dieldrin blank levels ranged from 0.004 to 0.33 ppb ($\bar{x} = 0.11$ ppb). Thus the apparent dieldrin peak in many samples may represent blank levels.

Fish collected from Coos Bay showed relatively low levels of DDE and PCB's (Table 3). PCB chromatograms resembled those found in the pink shrimp, i.e., Aroclor 1254 with two peaks missing; starry flounder chromatograms resembled those of Aroclor 1260.

Columbia River fish contained much higher levels of chlorinated hydrocarbons (Table 3), particularly PCB's (90-1160 ppb), than those found in Coos Bay fish. Starry flounder was the only species collected from both estuaries. Columbia River samples had six times the level of ΣDDT and 11 times the level of PCB's than had Coos Bay specimens. Interfering peaks prevented determination of dieldrin levels in these samples.

Summer-run steelhead from the Rogue River contained highest levels of DDE (97 ppb) and dieldrin (21 ppb) of all species sampled. In addition, 6 ppb chlordane and thiodan were found in several samples. PCB levels in Rogue River fish were lower than in Columbia River fish.

Results for mollusks are summarized in Table 4. The only chlorinated hydrocarbon pesticide found consistently throughout the sampling area was *p,p'*-DDE and, occasionally, *p,p'*-TDE and *p,p'*-DDT. Concentrations of DDT and related compounds were very low except in the Columbia River estuary. There were no significant seasonal variations. The Coos Bay *Mya* population had higher DDT residues (1.6-3.0 ppb) than had other clam populations from estuaries with small coastal mountain watersheds, i.e., Umpqua, Tillamook, and Yaquina. Only a single species, *Clinocardium nuttallii*, was analyzed from Netarts Bay; no chlorinated hydrocarbon compounds were detected.

Levels of DDT compounds in mollusks from the Columbia River were in marked contrast to those in mollusks from the other Oregon estuaries. Unfortunately, neither *Clinocardium* nor *Mya* was available from this system so comparisons must be made using different species. Except for a single sample of *Mytilus edulis* taken from the south jetty during the winter period, only *Anodonta* sp. and *Corbicula fluminea* were collected at this location. Levels of ΣDDT in *Anodonta* ranged from 14.9 ppb during the spring to 2 ppb in the fall. In contrast, ΣDDT in *Corbicula* ranged from

53 to 78 ppb. No marked seasonal variation was evident in this latter species either in ΣDDT or in the proportion of metabolites.

The only other pesticide detected in mollusks during the sampling period was dieldrin, which was present in all three Columbia River species analyzed during the winter and spring but was not detected in the summer and fall collections. Concentrations in mollusk tissues never exceeded 4 ppb.

During the first three sampling periods PCB's were found only in tissues of Columbia River bivalves. The PCB's found are believed to be a mixture of Aroclor 1254 and Aroclor 1260. *Corbicula* samples had consistently higher levels of PCB's than had Anodonta samples. In the former species concentrations ranged from 390 to 1,170 ppb. The highest level was found in the spring sampling period; levels did not exceed 570 ppb during the three remaining seasons. Levels of PCB's in *Anodonta* ranged from 160 ppb to levels

TABLE 4. *Chlorinated hydrocarbons in Oregon estuarine mollusks, 1972*

Species	Date	Concentration, μg/kg wet weight			
		p,p'-DDE	*p,p'*-TDE	*p,p'*-DDT	PCB'S
Columbia River					
Corbicula fluminea	2/24	21	20	12	570
	4/14	ND	ND	ND	1170
	7/28	35	28	15	390
	10/5	30	17	10	420
Anodonta	4/14	7	4	3	160
	7/28	7	2	1	35
	10/4	2	NS	NS	NS
Mytilus edulis	2/24	NS	NS	NS	44
Coos Bay					
Mya arenaria	1/25	0.6	0.8	2	NS
	4/17	0.7	NS	1.6 [1]	NS
	7/12	0.7	0.7	3	NS
	9/30	ND	NS	NS	26
Clinocardium nuttallii	1/25	NS	NS	NS	NS
	4/17	NS	NS	NS	NS
	7/12	0.3	NS	0.4	NS
	9/30	ND	NS	NS	NS
Tillamook					
Mya arenaria	1/14	NS	NS	NS	NS
	4/13	NS	NS	NS	NS
	7/9	0.2	NS	0.3	NS
	10/5	0.6	NS	NS	NS
Clinocardium nuttallii	1/14	NS	NS	NS	NS
	4/13	0.5	NS	0.4	NS
	7/9	0.1	NS	NS	NS
	10/5	0.6	NS	NS	NS
Umpqua					
Mya arenaria	1/13	0.6	<0.7	<0.5	NS
	1/13	0.6	NS	NS	NS
	4/17	1.0	0.5 [1]	0.4 [1]	NS
	7/11	0.2	0.3	NS	NS
	10/2	1.2	NS	NS	NS
Mytilus edulis	1/26	0.7	NS	NS	NS
	5/19	1.0	NS	NS	NS
	7/12	0.8	0.3	NS	NS
	10/2	1.8	NS	NS	NS
Yaquina					
Mya arenaria	1/24	0.2	NS	NS	NS
	4/15	0.5	NS	NS [1]	NS
	7/8	0.2	NS	NS	NS
	9/25	0.6	NS	NS	5
Clinocardium nuttallii	1/1	NS	NS	NS	NS
	4/15	0.2	0.5	NS	NS
	7/8	0.2	NS	NS	NS
	9/25	0.3	NS	NS	7
Blanks [2,3]	Winter	0.2	0.3	0.5	19
	Spring	0.2	0.2	0.4	9
	Summer	0.1	0.2	0.4	5
	Fall	0.2	0.2	0.4	1

NOTE: Values have been corrected for sea water blanks.
ND = no data because of interference with analytical process.
NS = residues not significant (less than twice the blank values).
[1] Confirmed by base hydrolysis.
[2] Values assume 30-g sample weight.
[3] Average of two blanks.

lower than those determined for blank samples. Like *Corbicula*, *Anodonta* displayed the highest PCB level in the spring. During the fall sampling period, low levels (5-7 ppb) of PCB's were detected in the two bivalve species from Yaquina Bay, and 26 ppb PCB's were found in the Coos Bay *Mya* population.

Discussion

Relative concentrations of DDT and PCB compounds differed in the three offshore species. Residues of ΣDDT were 3 ppb in both euphausiids and pink shrimp, but PCB levels differed between the two species by a factor of three: 7.5 versus 25 ppb, respectively (Table 3). Levels of DDT in flatfish were three times higher than in euphausiids and pink shrimp, but PCB levels were similar to those in the pink shrimp. Considering that all three species have nearly the same lipid content, approximately 2 percent, other factors probably account for these differences. Both euphausiids and pink shrimp feed on zooplankton and smaller animals. Euphausiids are found in the water column, however, and pink shrimp are found near the ocean floor. Another possible explanation for the difference between these levels is that the two species were collected from different geographic locations. Most euphausiids were collected west of Newport (station 7); pink shrimp were collected farther north at stations 2 and 5.

DDT levels reported by Giam et al. (*2*) for the Gulf of Mexico are considerably higher. DDT levels for Gulf shrimp (family Panaeidae) ranged from 33 to 165 ppb; PCB chromatograms lacked sufficient resemblance to an Aroclor formulation for quantitation. DDT levels in Gulf fish were also much higher than those from the study reported here, but PCB levels were comparable.

Atlantic Ocean levels (*1*) are similar to those reported from the present study of the Northeast Pacific. Icelandic shrimp (*Pandalus borealis*) contained 1 and 18 ppb DDT and PCB compounds, respectively.

In 1968 Stout (*12*) reported pesticide residues in fish and shellfish in the Northeast Pacific. Residues in hake collected along the Oregon/Washington coast ranged from 115 to 285 ppb total DDT; DDE represented only 26-36 percent of the total DDT residue. Some PCB interference may have accounted for higher DDD and DDT residues. In the authors' 1972 collections, DDE often represented the major portion of the total DDT residue; Columbia River collections, which showed signs of recent DDT contamination, were the exception.

Little is known about biological effects of PCB's on the marine environment. Duke et al. (*13*) exposed shrimp (*Penaeus duorarum*) to 5 ppb Aroclor 1254 in sea water for 20 days. Shrimp that died after 10 days had only 1,600 ppb PCB's; those living after 20 days had 3,300 ppb PCB's. Thus mortality probably was not caused by PCB poisoning. If 1,600 ppb is taken as a toxic residue level for shrimp, then pink shrimp (*Pandalus jordani*) in the Northeast Pacific contain only 1/60 the toxic residue level. Similar studies for DDE were not located.

DDE and PCB levels in the Coos Bay fish were slightly less than those found in the offshore flatfish collections (5 vs. 9 ppb DDE and 22 vs. 29 ppb PCB's, respectively).

Traces of chlordane and thiodan found in the steelhead may have originated from agricultural use in Medford, Oreg., a fruit-growing area. Only minor quantities of these chemicals are presently being applied in this area. Dieldrin found in those collections may have originated in the Rogue River, although dieldrin was also found in a few offshore collections.

PCB chromatograms of shrimp samples indicate selected metabolism of some isomers. All shrimp species had very low peaks for isomers 21 and 23 although euphausiids contained the expected ratio of isomers. The lower quantities of isomers 21 and 23 in flatfish may be a result of their feeding on pink shrimp. The larger fish, herring, salpa, and steelhead, had chromatograms closely resembling Aroclor 1254. PCB chromatograms of common murres collected in this area were very similar to the shrimp chromatogram (*11*). It is significant that three of the four fish collected in the Coos Bay estuary had PCB patterns closely resembling Aroclor 1254. The pattern of the fourth, a starry flounder, resembled Aroclor 1260.

Except for the Columbia River collections, organochlorine residues reported here for Oregon estuarine mollusks are consistently lower than those reported for mollusks from many other coastal States. Coos Bay mollusks had higher ΣDDT residues than had mollusks from the small coastal drainage estuaries, but even these did not exceed 5 ppb. In contrast, a very high percentage of mollusks sampled in other States contained between 11 and 100 ppb ΣDDT; significant numbers contained even more than 100 ppb (*5*).

Residues found in Columbia River *Corbicula* (53-78 ppb) more closely paralleled those reported in other States, but *Anodonta* collected in the same area contained less than 15 ppb ΣDDT. In general, a higher level of DDT tissue residues would be anticipated in Columbia River mollusks considering the enormous area of agricultural land drained by this river system. However, high levels of ΣDDT found in *Corbicula* may also be a result of the extraordinary ability of this species to accumulate organochlorine compounds.

Like DDT, PCB's accumulated far more heavily in *Corbicula* than in the other mollusks examined. The higher levels of PCB's in the Columbia River fish suggest that PCB contamination of the Columbia River greatly exceeded that of adjacent coastal waters.

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Residues of Organochlorine Pesticides and Polychlorinated Biphenyls and Autopsy Data for Bald Eagles, 1971-72 [1]

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ABSTRACT

Thirty-seven bald eagles found sick or dead in 18 States during 1971-72 were analyzed for organochlorine pesticides and polychlorinated biphenyls (PCB's). DDE and PCB's were detected in all bald eagle carcasses; 30 carcasses contained DDD and 28 contained dieldrin. Four eagles contained possibly lethal levels of dieldrin and nine eagles had been poisoned by thallium. Autopsies revealed that illegal shooting was the most common cause of mortality. Since 1964 when data were first collected, 8 of the 17 eagles obtained from Maryland, Virginia, South Carolina, and Florida possibly died from dieldrin poisoning; all four specimens from Maryland and Virginia were from the Chesapeake Bay Tidewater area.

Introduction

The purpose of this paper is to report and evaluate residue and autopsy data on bald eagles *(Haliaeetus leucocephalus)* collected in 1971 and 1972. Data for specimens collected in 1964 through 1970 have been previously reported (*1-3*).

Sampling

Bald eagles found dead or moribund in the field are collected by Federal, State, and private cooperators, packed in dry ice, and shipped air express to the Patuxent Wildlife Research Center in Laurel, Md., where they are stored intact in plastic bags at −25° C. Thus sampling for the present study was not systematic because of the relatively low population and protected status of these birds. Table 1 shows the collection areas of the 37 birds analyzed; 25 birds were collected in 1971 and 12 in 1972. Decomposed specimens were not analyzed.

TABLE 1. *Distribution of eagles collected by State and year of death, 1971-72*

State	No. Eagles Collected	
	1971	1972
California	1	
Florida	1	
Illinois	4	
Indiana		1
Iowa	1	
Maine		1
Michigan		1
Minnesota		2
Missouri	4	1
New Mexico		1
New York		1
Ohio	1	
South Carolina	1	
Texas		1
Utah	2	
Virginia	1	2
Wisconsin	1	
Wyoming	8	1
TOTAL	25	12

Autopsy and Analytical Procedures

Procedures for autopsy followed those reported previously by Belisle et al. (*3*). After removal of the skin, feet, wings, liver, and gastrointestinal tract, the carcass was ground and homogenized in a Hobart food cutter. A 10-g aliquot of the carcass and the entire brain were

[1] Patuxent Wildlife Research Center, Fish and Wildlife Service, U.S. Department of Interior, Laurel, Md. 20811.

mixed separately with anhydrous sodium sulfate in a blendor and extracted for 7 hours with hexane in a Soxhlet apparatus. Extracts were evaporated, lipid weights were determined, and the extracts were redissolved in 20 ml hexane. A 10-ml aliquot of extract containing not more than 0.5 g lipid was cleaned on a florisil column.

The florisil had been washed and recalcined at 675° C according to Hall's method (*4*) and partly deactivated with 1.0-1.5 percent water to permit the elution of dieldrin with the other pesticides. Approximately 21 g of the treated florisil was placed in each 2-by-20-cm column with a 250-ml reservoir and topped with 1 cm anhydrous sodium sulfate. Columns were prewashed with 50 ml hexane and the extract was eluted with 200 ml 6 percent ethyl ether in hexane.

The florisil eluate was concentrated to 5 ml and a 4-ml aliquot was placed on a silicic acid column to separate pesticides from PCB's. Armour and Burke's separation method (*5*) was used with the following modifications: the silicic acid, Mallinckrodt Silicar CC-4, was heated at 130° C for 24 hours in a pan covered with aluminum foil containing a few pinholes; celite and air pressure were eliminated; and the petroleum ether eluate was collected in two separate fractions of 100 ml and 300 ml followed by 200 ml of the polar eluate. The adsorbent usually was deactivated with 3 percent water, and the flask was sealed with paraffin tape, shaken for 3 hours on a reciprocating shaker, and allowed to equilibrate for 24 hours before use. The amount of water was adjusted by running standards to assure that all the DDE was in the second fraction.

Using this procedure, hexachlorobenzene (HCB) and mirex were collected in the first 100 ml petroleum ether, PCB's and DDE were in the second fraction, and the remaining pesticides were in the polar eluate. Silicar CC-4 did not require celite or air pressure to maintain the specified flow rate. Covering the pan with aluminum foil during the heating process eliminated certain interfering background peaks.

Samples were analyzed with a Hewlett-Packard 5753 gas-liquid chromatograph equipped with a Ni^{63} detector, automatic sampler, digital integrator, and a 4 percent SE-30/6 percent QF-1 column at 190° C. The flow rate of 5 percent methane in argon was 60 ml/min for columns and 40 ml/min for purge. DDE was quantitated by peak height to avoid possible errors from PCB interference; other pesticides were measured by digital integration of area, and PCB's were estimated by comparing total peak area with Aroclor 1254 or 1260.

Residues in 15 specimens (40 percent) were positively identified with an LKB gas-liquid chromatograph/mass spectrometer (GLC/MS). Operating procedures have been described (*3*) except that a 1 percent SE-30 column was temperature-programmed. Program rate was 2° C/min; initial temperature was 135° C, rising to a maximum of 220° C.

Average recoveries from spiked mallard carcass tissue were: DDE, 96 percent; DDD, 103 percent; DDT, 110 percent; dieldrin, 101 percent; heptachlor epoxide, 104 percent; mirex, 106 percent; oxychlordane, 98 percent; *cis*-chlordane, 100 percent; *cis*-nonachlor, 98 percent; HCB, 69 percent; and Aroclor 1254, 101 percent. Residue levels for eagle samples were not corrected for recovery. The lower limit of sensitivity was 0.05 ppm; residue levels less than 0.05 ppm were not reported "trace" as in the previous reports.

Samples were not analyzed for oxychlordane, *cis*-chlordane, *cis*-nonachlor, or HCB in 1971. GLC/MS analyses using temperature programming revealed both *cis*-chlordane and *trans*-nonachlor. In one sample, only *trans*-nonachlor was detected. Authors were unable to obtain a GLC column for the electron-capture detector that would separate both compounds without interference from another pesticide. The peak was quantitated as *cis*-chlordane because standards of these compounds have the same detector response.

Thallium levels in eagle kidneys were determined by flame atomic absorption using the method described by Curry et al. (*6*) except that a sampling boat was not used. The lower limit of sensitivity was 2.0 ppm.

Results and Discussion

RESIDUES

Table 2 summarizes residues of organochlorine pesticides and PCB's in 37 bald eagle carcasses and brains; all data are reported on a wet-weight basis. All carcasses contained PCB and DDE residues, 30 contained DDD, and 28 contained dieldrin.

Four specimens had concentrations of dieldrin in the brain within the range known to have caused death by dieldrin. Table 3 shows dieldrin levels in the brains of these specimens to range from 4.0 to 7.8 ppm. Stickel et al. (*7*) concluded from an experimental study on Japanese quail *(Coturnix coturnix)* and from residues in brains of several kinds of animals found dead in the field following heavy dieldrin treatments that a concentration of 4-5 ppm indicated that the animal was in the danger zone. Linder et al. (*8*) concluded from studies of capsule-dosed pheasants that a level of 3-4 ppm, or greater, of dieldrin in the brain indicates death by dieldrin.

During the 1964-72 period, 190 eagles were analyzed; 19 (10 percent) of these specimens were suspected cases of dieldrin poisoning. The incidence of dieldrin poisoning is high, particularly among specimens from Maryland, Virginia, South Carolina, and Florida. Of the 17 eagles collected (3 from Md., 4 from Va., 4 from S.C., and 6 from Fla.), 8 (47 percent) were possible victims

TABLE 2. *Pesticide and PCB residues in 37 bald eagles, 1971-72*

Compound	Year	Residues, ppm wet weight					
		Carcass			Brain		
		No. specimens [1]	Median [1]	Range	No. specimens [1]	Median [1]	Range
p,p'-DDE	1971	25	5.7	0.77- 210.0	25	0.95	0.07- 89.0
	1972	12	11.0	0.83- 110.0	12	3.3	0.14- 55.0
p,p'-DDD	1971	18	0.40	0.10- 33.0	9	0.19	0.05- 9.9
	1972	12	0.54	0.14- 18.0	8	0.35	0.06- 2.9
p,p'-DDT	1971	4	0.33	0.26- 3.2	2	0.08	0.05- 0.11
	1972	5	0.42	0.12- 0.94	1	0.34	-
Dieldrin	1971	16	0.72	0.10- 33.0	12	0.30	0.05- 7.8
	1972	12	0.65	0.14- 12.0	8	0.61	0.23- 4.6
Heptachlor epoxide	1971	9	0.36	0.06- 5.5	6	0.18	0.05- 1.5
	1972	5	0.72	0.06- 2.7	3	0.35	0.11- 1.7
Mirex	1971	10	0.30	0.10- 1.3	4	0.10	0.10- 0.13
	1972	6	0.26	0.06- 0.60	2	0.13	0.11- 0.13
Oxychlordane	1972	6	0.34	0.18- 1.4	6	0.09	0.05- 0.34
cis-Chlordane [2]	1972	10	0.30	0.11- 7.4	7	0.11	0.05- 1.7
cis-Nonachlor	1972	6	0.29	0.07- 0.95	2	0.59	0.19- 0.98
Hexachlorobenzene	1972	4	0.30	0.11- 0.50	4	0.14	0.05- 0.18
PCB's	1971	25	8.6	0.30- 290.0	24	1.1	0.10-150.0
	1972	12	26.0	0.60-1200.0	11	16.0	0.65-190.0

[1] Number of specimens containing residues; the median is based on this number.
[2] And/or *trans*-nonachlor.

TABLE 3. *Data on four suspected cases of dieldrin poisoning of adult eagles, 1971-72*

State	Year	Sex	Dieldrin residue in brain, ppm	Autopsy findings [1]
Iowa	1971	F	7.8	Open, no fat deposits
South Carolina	1971	M	6.5	Emaciation
Virginia	1971	M	4.0	Open, no fat deposits
Virginia	1972	F	4.6	Open, no fat deposits

[1] Open = no diagnosis could be made on the basis of autopsy findings.

of dieldrin poisoning. Two eagles suspected of dieldrin poisoning were collected from each State. It is of special concern that all four eagles from Maryland and Virginia suspected of dieldrin poisoning were from the Chesapeake Bay tidewater area. No specimens were collected from North Carolina or Georgia.

A large group of bald and golden eagles found dead near Casper, Wyo., in May 1971 were suspected of having been killed by ingestion of poisoned bait. Specimens were analyzed for cyanide, strychnine, sodium fluoroacetate (1080), and thallium. Residues of thallium in the kidneys ranged from 14 to 59 ppm. Eight of the eleven bald eagles from this group which were suitable for residue analysis are included in this report. In an eagle from Utah, the kidney contained 63 ppm thallium. It was concluded from autopsies and chemical analyses that these eagles had died from thallium ingestion.

AUTOPSY

Results of the autopsies are summarized in Table 4. Illegal shooting, the most common cause of mortality, was responsible for the death of 35 percent of the eagles.

Three eagles suspected of dieldrin poisoning were in good flesh but lacked deposits of fat. The other bird listed as emaciated showed marked atrophy of the pectoral and leg muscles, and had no fat deposits.

TABLE 4. *Probable causes of bald eagle mortality, 1971-72*

Cause of Death	No. Eagles
Shooting	13
Thallium poisoning	9
Dieldrin poisoning	4
Nephrosis	1
Drowning	1
Drowning; coccidiosis	1
Coccidiosis	1
Strychnine poisoning	1[1]
Impact	1
Electrocution	1
Open [2]	4
TOTAL	37

[1] Specimen from New Mexico; analyzed for strychnine by Denver Wildlife Research Center, Denver, Colo.
[2] No diagnosis could be made on the basis of autopsy findings or chemical analysis.

Coccidiosis caused by *Isospora* sp. was apparently responsible for the death of an eagle from Minnesota. It was a contributing factor in the death of an emaciated adult female found downstream from Flaming Gorge Dam, Utah; immediate cause of her death was reported as drowning.

Drowning was also the immediate cause of the death of an adult female eagle from Michigan that had high levels of DDE (55 ppm) and PCB's (190 ppm) in the brain. The eagle, which had been observed perched on a snag over the water, suddenly fell into the water and drowned. No injuries were found, and both aerobic and anaerobic cultures of liver, heart blood, and intestinal tract failed to reveal any pathogenic bacteria.

The thallium-poisoned eagles usually had normal amounts of adipose tissues, were in good flesh, and had no gross lesions except congestion of vessels overlying the cerebellum. Microscopically, there were no acid-fast intranuclear inclusion bodies which sometimes indicate lead poisoning, and examination of the kidney sections stained by the Pritschow technique proved to be equivocal.

An adult female eagle from Missouri had a shattered lower left leg, the result of an earlier gunshot wound. She had valvular endocarditis, probably the result of a secondary bacterial infection from the leg wound. *Escherichia coli* appeared in the heart blood but not in the lungs and liver; thus the cause of death is listed as shooting (Table 4).

Conclusion

Bald eagles are subject to a wide variety of environmental insults, including infectious agents, chemical pollutants, and human-related trauma. Levels of pesticides and PCB's in eagles continue to be high, reflecting widespread contamination by these compounds.

Acknowledgments

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Mercury Concentrations in Fish, North Atlantic Offshore Waters—1971

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ABSTRACT

Mercury concentrations were determined in muscle and liver of 41 species of fish and a limited number of plankton, sediment, and invertebrate samples collected from North Atlantic offshore waters in 1971. The average mercury concentration in fish muscle was 0.154 ppm with a standard deviation of 0.124. Invertebrate samples had mercury concentrations which were generally less than 0.1 ppm. In a single lobster sample, however, 0.31 ppm mercury was found in the tail muscle and there was 0.60 ppm in the liver. Mercury levels in all 9 plankton and 10 sediment samples taken were less than 0.05 ppm.

Introduction

Recently, much has been published about mercury in freshwater lakes in Japan, Sweden, Canada, and the Great Lakes area of the United States. Mercury discharged into these waters was found to accumulate in tissues of fish and other organisms to levels that in certain species were considered potentially dangerous to human health (*1*).

This mercury was traced to many industrial and domestic uses, such as the manufacture of sodium hydroxide and chloralkali plants, paper manufacturing, plastics production, and application of fungicides to control yeast and mold growth on grass and in pulp mills.

The degree and source of mercury contamination of freshwater fish and waters were readily established. Levels in marine fish and waters, however, were not so easily determined. The Food and Drug Administration (FDA), U.S. Department of Health, Education, and Welfare, conducted a survey of mercury levels in several species of both domestic and foreign marine fisheries products and found that certain species of tuna and swordfish contained mercury above the 0.5 ppm action level, the maximum allowable concentration in fish intended for sale.

As a result of these findings, a program was initiated within the National Marine Fisheries Service, U.S. Department of Commerce, to determine mercury levels in other marine fish as part of an overall program on the effects of chemical contamination of living marine resources. The present paper reports on part of this program, a survey of mercury concentrations in groundfish collected from U.S. waters of the North Atlantic Ocean.

Experimental Methods

SAMPLE COLLECTION

Fish and invertebrates were collected by otter trawl during the annual assessment of groundfish stocks conducted by the National Marine Fisheries Service, Northeast Fisheries Center, Woods Hole, Mass. After the catch, fish and invertebrates were sorted and dissected aboard the vessel. Livers and a 1-inch-thick steak immediately posterior to the head were taken from each fish. Invertebrate samples varied: whole squid were analyzed although scallop samples were composed of only the edible muscle and lobster samples consisted of the digestive diverticula and tail muscle.

Bottom sediments were obtained from selected areas with a Smith-McIntyre sampler. Samples were removed for analysis with a plastic tube 1½ inch in diameter and 6 inches long, which was inserted into the bottom sediment, capped, and frozen.

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Approximate geographic sampling areas are shown in Figure 1. Common and scientific names of fish and invertebrates obtained in the survey are presented in Table 1.

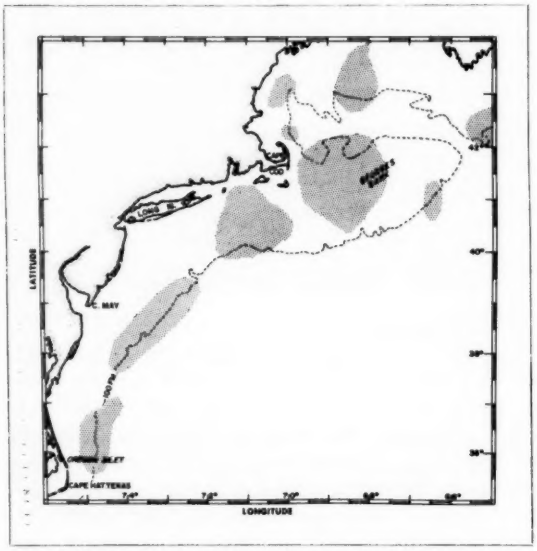

FIGURE 1. *Collection sites of fish sampled for mercury concentrations, North Atlantic offshore waters—1971*

TABLE 1. *Fish sampled for mercury concentrations, North Atlantic offshore waters—1971*

COMMON NAME	SCIENTIFIC NAME
American plaice	*Hippoglossoides platessoides*
American shad	*Alosa sapidissima*
Angel shark	*Squalus dumerili*
Atlantic cod	*Gadus morhua*
Atlantic herring	*Clupea harengus harengus*
Atlantic mackerel	*Scomber scombrus*
Atlantic wolffish	*Anarhichas lupus*
Beardfish	*Polymixia lowei*
Blackbelly rosefish	*Helicolenus dactylopterus*
Black sea bass	*Centropristis striata*
Butterfish	*Peprilus triacanthus*
Cusk	*Brosme brosme*
Daubed shanny	*Lumpenus maculatus*
Fawn cusk-eel	*Lepophidium cervinum*
Fourspot flounder	*Paralichthys oblongus*
Gulf Stream flounder	*Citharichthys arctifrons*
Haddock	*Melanogrammus aeglefinus*
Lanternfish	Unclassified
Little skate	*Raja erinacea*
Longhorn sculpin	*Myoxocephalus octodecemspinosus*
Mailed sculpin	*Triglops nybelini*
Northern searobin	*Prionotus carolinus*
Ocean pout	*Macrozoarces americanus*
Pollock	*Pollachius virens*
Redfish	*Sebastes marinus*
Red hake	*Urophycis chuss*
Round herring	*Etrumeus teres*
Silver hake	*Merluccius bilinearis*
Spiny dogfish	*Squalus acanthias*
Spot	*Leiostomus xanthurus*
Striped searobin	*Prionotus evolans*
Thorny skate	*Raja radiata*
White hake	*Urophycis tenuis*
Windowpane flounder	*Scophthalmus aquosus*
Winter flounder	*Pseudopleuronectes americanus*
Winter skate	*Raja binoculata*
Witch flounder	*Glyptocephalus cynoglossus*
Yellowtail flounder	*Limanda ferruginea*

SAMPLE PREPARATION FOR CHEMICAL ANALYSIS

At the laboratory fish steaks were thawed, skinned, and boned. Muscle and liver tissues of 5-10 fish from each station were combined into single composite samples for mercury analysis although some muscle and liver tissues were also analyzed individually. Invertebrates were pooled into composites of 10 animals per station. Samples were homogenized in an electric blendor composed of stainless steel blades and a glass jar.

Plankton samples were processed in a Vir-Tis model 10-100 freeze-drier for 48 hours. Plankton data are reported on a wet-weight basis. Entire sediment samples about 3-5 inches deep and 1½ inches in diameter were thoroughly mixed by hand in a plastic bag prior to analysis.

MERCURY ANALYSIS

Samples were analyzed according to the procedure of the Division of Laboratories, Ontario Ministry of the Environment (*2*). Plankton samples and ground flesh or liver samples ranging from 0.1 to 0.5 g were weighed into 30-ml Kjeldahl flasks. Ten ml 4:1 reagent grade concentrated sulfuric acid:nitric acid were added and the sample was shaken in a 50°-60° C water bath. Digestion was considered complete after 1½-2 hours when a clear solution was obtained. Flasks and samples were cooled at room temperature for 1 hour and placed in ice while 15 ml 6 percent potassium permanganate was slowly added to each sample. After addition of permanganate, samples were left overnight at room temperature.

A 20 percent sulfuric acid solution was added and samples were transferred to glass washing bottles equipped with fritted stems. Twenty ml reductant consisting of 100 ml concentrated sulfuric acid, 15 g sodium chloride, 30 g hyroxylamine sulfate, and 60 g stannous sulfate made up to 1,000 ml with distilled water were added to the sample and stirred for 1 minute. Mercury was swept by air through a 2.5-cm cell mounted in the light path of a Perkin Elmer model 305 atomic absorption spectrophotometer. Air flow rate was adjusted to give about 60 percent recorder response for 0.3 μg mercury. Peak heights of sample recorder response were compared to those of standards carried through the same digestion procedure described above for quantitation.

The method was checked for accuracy by comparing its results with those obtained by other procedures. A sample of oil-packed yellow fin tuna prepared by the National Canners Association was analyzed for mercury residues by nine laboratories using a variety of methods. The average mercury level obtained was 0.86 ppm; range was 0.80-1.02 ppm. The tuna sample studied at this laboratory was routinely analyzed with small batches of samples taken in the present study. An average value of 0.90 ppm with a relative standard deviation of 17.94 percent was obtained for 39 replicate determinations.

The procedure for mercury analysis of sediment samples was obtained from the Chemistry Laboratory Manual—Bottom Sediments, December 1969, compiled by the Great Lakes Region Committee on Analytical Methods, Federal Water Quality Administration, a predecessor of the U.S. Environmental Protection Agency. Samples ranging between 0.2 and 0.5 g were weighed into 125-ml Erlenmeyer flasks and 10 ml distilled water and 5 ml of 1:3 lactic acid:HCl (aqua regia) were added. Samples were heated for 2 minutes in a 95° C water bath and cooled in tap water for 10-15 minutes. Fifty ml distilled water and 15 ml of a 6 percent potassium permanganate solution were added to each flask. Then samples were reduced and analyzed by atomic absorption spectrophotometry as described for fish tissue except that sediment samples were analyzed the day they were digested.

Percent recoveries of $HgCl_2$ added to fish muscles and sediment prior to digestion are given in Table 2. Mercury was added to fish muscle before any acids or other reagents and was not allowed to equilibrate with muscle prior to addition of acid. Mercury added to sediment was allowed to stand for 2 hours prior to addition of water and acid. Sensitivity was about 0.05 ppm.

TABLE 2. *Percent recovery of mercury from fish and sediment, North Atlantic offshore waters—1971*

Species	Mercury Added, μg [1]	No. Replicates Analyzed	Recovery, %	
			Range	Average
Fishmeal	0.05	6	81.9-118.1	95.4
Swordfish	0.1	6	100.7-127.9	113.7
Yellow perch	0.2	6	80.9-108.0	96.9
Carp	0.2	6	89.7-102.3	95.1
Yellowfin tuna	0.3	5	95.4-105.1	98.6
Skipjack tuna	0.5	5	103.1-111.9	108.2
Sediment	0.3	6	105.0-119.0	111.5

[1] $HgCl_2$ used for mercury addition.

Results

Mercury concentrations among individual specimens were similar for most fish species although variations among individuals were observed for cusk and spiny dogfish (Table 3). One cusk collection had mercury concentrations ranging from 0.14 to 1.33 ppm in the liver and from 0.15 to 0.59 ppm in the muscle. In a second collection, variation among individuals was less: residues ranged from 0.16 to 0.34 ppm in the muscle and from 0.13 to 0.40 ppm in the liver.

Variation among four of six collections of spiny dogfish individuals was substantial. Mercury levels in these samples were: 0.20-0.61 ppm, 0.35-0.69 ppm, 0.35-0.93 ppm, and 0.22-0.65 ppm. Mercury content among individuals in the other two collections ranged only from 0.17 to 0.29 ppm and from 0.14 to 0.29 ppm.

MERCURY LEVELS AND FEEDING HABITS

In an attempt to correlate mercury concentrations with feeding habits, fish were grouped according to similar feeding patterns. The majority collected were bottom-feeders, others were primarily pelagic and plankton feeders and a few species could not be grouped into a particular feeding habit and were listed as miscellaneous. Feeding habits and mercury concentrations did not correlate (Table 4).

MUSCLE

Highest levels of mercury in fish muscle were found in cusk, spiny dogfish, northern searobin, and striped searobin (Table 5). The highest mercury concentrations in these samples were 0.49, 0.53, 0.35, and 0.35 ppm, respectively. The 36 other fish species had mercury levels in muscle that were less than 0.30 ppm. Mercury levels in all fish muscle sample averaged 0.154 ± 0.124 ppm.

LIVER

Highest mercury levels in fish livers were detected in blackbelly rosefish, cusk, northern searobin, and American shad (Table 5). The highest mercury levels in these samples were 0.40, 0.83, 0.56, and 0.67 ppm, respectively. The other 36 species of fish had mercury levels below 0.30 ppm in the liver (Table 5). Concentrations in livers of all fish examined averaged 0.164 ± 0.157 ppm.

MUSCLE AND LIVER

Mercury residues in fish samples averaged 0.154 ppm in muscle and 0.164 ppm in liver. Investigators have shown that liver accumulates metals to a greater extent than do most other tissues and organs (*3-5*). Data in this study, however, reflected an important difference: similar concentrations of mercury occurred in muscle and liver for most species examined. Exceptions to this rule were spiny dogfish, blackbelly rosefish, American shad, and Atlantic herring. Levels in spiny dogfish were two to three times higher in muscle than in liver, whereas in the other species mercury concentrations were greatest in the liver. Ratios of mercury levels in liver to those in muscle for other species were blackbelly rosefish, 2:1; American shad, 13:1; and Atlantic herring, 5:1.

PLANKTON, SEDIMENT, AND INVERTEBRATES

Mercury levels in all 9 plankton and 10 sediment samples were less than 0.05 ppm (Table 6). Pandallis shrimp, scallops, and squid generally had mercury levels less than 0.05 ppm (Table 7). However, the mercury levels of one squid sample and one shrimp sample were 0.06 ppm and 0.09 ppm, respectively. The single lobster sample obtained had mercury levels of 0.31 ppm in the tail meat and 0.60 ppm in liver (Table 7).

Discussion

This study measures total mercury: organic and inorganic forms. Methylmercury is considered more toxic to humans than are inorganic mercury salts and thus its occurrence in fish is of more toxicological significance

TABLE 3. *Mercury concentrations in individual fish samples, North Atlantic offshore waters—1971*

Species	Tissue	No. Fish Analyzed	Mercury Content, ppm wet weight		
			Range	Average	Standard Deviation
American dab	muscle	5	0.03-0.16	0.08	0.064
Atlantic cod	muscle	4	0.10-0.15	0.14	0.081
	liver	4	0.08-0.15	0.12	0.017
	muscle	5	0.10-0.38	0.24	0.135
	liver	5	0.10-0.36	0.18	0.109
Atlantic herring	liver	5	0.20-0.44	0.26	0.103
Atlantic wolffish	muscle	5	0.08-0.25	0.16	0.063
Blackbelly rosefish	muscle	5	0.12-0.31	0.22	0.095
Cusk	muscle	6	0.16-0.34	0.27	0.089
	liver	6	0.13-0.40	0.24	0.095
	muscle	6	0.15-0.59	0.41	0.189
	liver	6	0.14-1.33	0.65	0.440
Fourspotted flounder	muscle	5	0.09-0.19	0.16	0.031
Haddock	liver	5	0.05-0.09	0.06	0.014
Little skate	muscle	5	0.05-0.18	0.12	0.060
	liver	5	0.06-0.15	0.10	0.033
Mackerel	muscle	5	0.07-0.10	0.08	0.016
Pollock	muscle	5	0.06-0.12	0.10	0.029
	liver	5	0.05-0.09	0.06	0.016
Redfish	muscle	6	0.15-0.29	0.20	0.050
Red hake	muscle	5	0.05-0.09	0.06	0.000
Silver hake	muscle	5	0.06-0.14	0.09	0.034
Spiny dogfish	muscle	4	0.17-0.29	0.23	0.052
	muscle	10	0.20-0.61	0.34	0.160
	muscle	5	0.35-0.69	0.54	0.135
	muscle	5	0.35-0.93	0.58	0.270
	muscle	5	0.22-0.65	0.44	0.154
	muscle	5	0.14-0.29	0.18	0.064
Thorny skate	muscle	5	0.11-0.41	0.19	0.116
	muscle	4	0.15-0.36	0.26	0.113
	liver	4	0.12-0.17	0.15	0.033
White hake	muscle	5	0.08-0.17	0.12	0.037
	liver	5	0.05-0.18	0.12	0.055
Windowpane flounder	muscle	5	0.06-0.18	0.10	0.045
Winter flounder	muscle	5	0.06-0.12	0.08	0.022
	liver	5	0.12-0.26	0.18	0.044
Witch flounder	muscle	5	0.08-0.11	0.09	0.000
	liver	5	0.07-0.17	0.12	0.045
Yellowtail flounder	muscle	5	0.07-0.13	0.10	0.022

TABLE 4. *Mercury concentrations in fish grouped according to feeding habits, North American offshore waters—1971*

Fish	No. Collections Analyzed [1]	Mercury Content, ppm wet weight			
		Muscle		Liver	
		Range	Average	Range	Average
Bottom Feeders					
American dab	2	0.06-0.08	0.07	0.11-0.14	0.13
Atlantic cod	2	0.14-0.25	0.20	0.11-0.20	0.16
Atlantic wolffish	2	<0.05-0.15	0.08	<0.05-0.06	<0.05
Blackbelly rosefish	1	0.22	0.22	0.40	0.40
Black sea bass	1	0.08	0.08	0.18	0.18
Cusk	4	0.15-0.49	0.31	0.14-0.83	0.42
Fourspot flounder	2	0.16	0.16	0.23-0.27	0.25
Gulf Stream flounder	2	0.05	0.05	ND	ND
Haddock	2	<0.05-0.09	0.06	<0.05	<0.05
Little skate	2	0.13-0.16	0.15	0.10-0.23	0.17
Longhorn sculpin	2	0.08-0.09	0.09	0.09-0.16	0.13
Ocean pout	2	<0.05-0.11	0.07	<0.05-0.09	0.06
Red hake	2	<0.05-0.05	<0.05	<0.05-0.08	0.06
Striped searobin	1	0.35	0.35	0.38	0.38
Thorny skate	2	0.21-0.26	0.24	0.09-0.15	0.12
White hake	2	0.10-0.12	0.11	0.12-0.16	0.14
Windowpane flounder	1	0.10	0.10	0.12	0.12
Winter flounder	3	0.06-0.14	0.09	0.07-0.18	0.11
Winter skate	1	0.15	0.15	0.18	0.18
Witch flounder	2	0.07-0.10	0.09	0.13-0.16	0.15
Yellowtail flounder	2	0.10-0.24	0.17	0.17-0.25	0.21
Pelagic Feeders					
Pollock	2	0.08-0.10	0.09	<0.05-0.06	<0.06
Redfish	2	0.10-0.20	0.15	0.15	0.15
Spot	1	<0.05	<0.05	<0.05	<0.05
Silver hake	1	0.09	0.09	0.10	0.10
Plankton Feeders					
American shad	1	0.05	0.05	0.67	0.67
Atlantic herring	2	<0.05-0.09	0.06	0.26-0.28	0.27
Mackerel	1	0.08	0.08	ND	ND
Miscellaneous					
Angel shark	1	0.08	0.08	<0.05	<0.05
Cusk-eel	1	0.11	0.11	0.19	0.12
Spiny dogfish	8	0.07-0.53	0.32	<0.05-0.19	0.10

NOTE: ND = no data.
[1] Each collection includes 6-10 animals.

than is the occurrence of total mercury. Reports conflict concerning methylmercury:total mercury ratios in fish. Japanese investigators report that methylmercury does not exceed about 15 percent of the total mercury in fish whereas Swedish scientists have reported that these mercury concentrations are mostly methylmercury (*6*).

The Food and Drug Administration bases its administrative guideline of 0.5 ppm mercury in fish on total mercury content. Thus data presented here can be compared to the guideline. The only muscle samples in this survey that approached this level were those in cusk and spiny dogfish. Cusk measuring 61-67 cm had an average mercury content of 0.49 ppm. Levels in three groups of spiny dogfish muscle averaged 0.44, 0.47, and 0.53 ppm, whereas levels in five other groups of dogfish ranged from 0.07 to 0.34 ppm. The average of 0.154 ± 0.124 ppm for all fish muscle samples shows most fish well below the guideline level.

With the exception of cusk and spiny dogfish, fish examined in this study do not have abnormal mercury concentrations in relation to levels in marine fish reported by other investigators. Twelve of 15 species of Oregon groundfish had mercury levels in the range of 0.08-0.24 ppm. Yellow rockfish, lingcod, and spiny dogfish had average levels of 0.37, 0.35, and 0.60 ppm, respectively (*7*). The latter concentration was higher than the level reported for spiny dogfish in the present study.

Of about 21 species of marine fish and shellfish, only swordfish consistently had mercury levels at or over 0.5 ppm in an FDA study of marine and freshwater fish (*8*). Three thousand samples of canned tuna had an average mercury content of 0.25 ppm; less than 4 percent of these samples had levels over 0.5 ppm. The average mercury content of 19 other species of marine fish and shellfish was 0.3 ppm although most were below 0.1 ppm.

Mercury levels in fish landed in England and Wales have also been determined (*9*). Investigators there reported that mercury levels in fish from deep-water fishing areas averaged 0.06 ppm. Those caught away from coastal waters in the North Sea had an average concentration of 0.10 ppm whereas residues in fish caught in

TABLE 5. *Mercury concentrations in composite fish samples, North Atlantic offshore waters—1971*

Species	Sample Data					Mercury Content		
	Latitude	Longitude	Depth, fathoms [1]	Date	Length, cm [1]	Average, ppm wet weight Whole Animal Muscle		Liver
American dab	42° 48′	70° 38′	15-62	4-17	32-36	0.08		0.14
	41° 21′	68° 46′	28-62	10-19	ND	0.06		0.11
American shad	ND	ND	23-62	ND	ND	0.05		0.67
Angel shark	36° 11′	74° 48′	58-105	4-4	43-58	0.08		<0.05
Atlantic cod	41° 44′	69° 40′	58-115	10-18	ND	0.25		0.20
	42° 12′	70° 06′	15-62	11-4	54-61	0.14		0.11
Atlantic herring	41° 38′	68° 37′	28-62	10-18	ND	<0.05		0.26
	40° 23′	71° 02′	28-62	3-10	20	0.09		0.28
Atlantic wolffish	42° 42′	66° 07′	30-52	4-25	78-93	0.15		<0.05
	42° 55′	65° 01′	50-105	11-7	20-74	0.07		<0.05
Beardfish	37° 07′	74° 33′	58-105	4-5	13-14		0.06	
Blackbelly rosefish	39° 11′	72° 32′	95-200	10-7	16-34	0.22		0.40
Black sea bass	36° 50′	74° 42′	28-62	4-5	22-24	0.08		0.18
Butterfish	36° 37′	75° 20′	10-32	4-3	10		0.06	
Cusk	42° 46′	66° 37′	48-105	11-13	61-67	0.49		0.83
	42° 48′	70° 07′	55-130	4-18	44-68	0.27		0.23
Daubed shanny	43° 44′	69° 08′	5-62	4-21	8-12		0.05	
Fawn cusk-eel	40° 13′	71° 04′	58-105	10-8	25-29	0.11		0.19
Fourspot flounder	40° 24′	71° 57′	28-62	3-11	16-19	ND		0.27
	39° 09′	73° 12′	28-62	10-6	21-34	0.16		0.23
Gulf Stream flounder	38° 01′	74° 14′	28-62	10-5	6-12	0.05		ND
Haddock	41° 32′	69° 31′	10-35	10-18	ND	0.09		<0.05
	41° 01′	67° 06′	28-62	3-16	46-68	<0.05		<0.05
Lanternfish	41° 35′	61° 55′	95-200	10-22	ND		<0.05	
Little skate	40° 22′	68° 46′	28-62	10-16	ND	0.16		0.23
	40° 57′	71° 18′	13-33	3-30	45-50	0.13		0.10
Longhorn sculpin	40° 46′	68° 35′	10-34	10-17	ND	0.09		0.16
	41° 12′	71° 17′	13-33	3-10	25	0.08		0.09
Mackerel	39° 59′	71° 40′	28-62	3-11	30-35	0.08		ND
Mailed sculpin	43° 35′	66° 41′	50-115	4-23	8-13		<0.05	
Northern searobin	37° 53′	74° 41′	10-32	10-9	24-27	0.35		0.56
Ocean pout	40° 23′	73° 06′	11-32	3-31	45-55	0.11		0.09
	40° 55′	71° 63′	11-35	3-10	46	<0.05		<0.05
Pollock	42° 48′	66° 37′	48-105	4-24	51	0.08		<0.05
	41° 25′	69° 08′	58-115	10-19	ND	0.10		0.06
Redfish	41° 42′	69° 16′	58-115	3-24	38	0.20		0.15
	41° 57′	69° 02′	58-115	10-19	41-57	0.10		0.15
Red hake	40° 24′	71° 57′	28-62	3-11	25-30	<0.05		<0.05
	40° 33′	71° 57′	28-62	9-30	27-34	0.05		0.08
Round herring	40° 09′	73° 31′	11-32	10-4	15-16		<0.05	
Silver hake	39° 55′	72° 05′	28-62	10-7	28-31	0.09		0.10
Spiny dogfish	42° 20′	65° 35′	48-105	4-29	68-75	0.23		0.07
	42° 17′	66° 34′	90-160	3-21	ND	0.32		0.06
	44° 16′	66° 51′	50-135	11-15	81-84	0.34		0.09
	40° 32′	71° 20′	28-62	3-11	37	0.07		<0.05
	36° 28′	75° 08′	10-32	4-3	80-92	0.53		0.19
	38° 14′	73° 44′	58-105	4-8	82-95	0.47		0.07
	40° 29′	72° 26′	11-32	10-1	60-86	0.44		0.15
	40° 43′	70° 26′	11-35	10-15	ND	0.18		0.12
Spot	37° 29′	75° 13′	10-32	10-10	27-29	<0.05		<0.05
Striped searobin	36° 00′	74° 53′	28-62	4-4	28-37	0.35		0.38
Thorny skate	42° 41′	66° 12′	30-52	4-25	60-93	0.21		0.09
	44° 07′	66° 35′	15-62	11-15	60-81	0.26		0.15
White hake	42° 44′	69° 40′	90-155	4-18	57-68	0.12		0.12
	42° 16′	70° 22′	15-62	11-3	27-72	0.10		0.16
Windowpane flounder	41° 09′	70° 47′	11-35	9-29	22-31	0.10		0.12
Winter flounder	40° 11′	67° 56′	15-62	4-21	22-46	0.07		0.08
	40° 42′	72° 31′	11-32	10-1	28-35	0.14		0.18
Winter skate	40° 46′	68° 35′	10-34	10-17	ND	0.15		0.18
Witch flounder	38° 13′	73° 51′	95-200	4-8	31-36	0.07		0.16
	40° 32′	70° 57′	28-62	10-8	42-50	0.10		0.13
Yellowtail flounder	40° 16′	73° 24′	11-32	3-31	28-32	0.24		0.17
	40° 29′	71° 49′	28-62	9-30	33-38	0.10		0.25

NOTE: ND = no data.
[1] Values reported in ranges only; depth range represents a bottom contour from which the fish was taken.

the Irish Sea were twice as high. The average level of about 0.15 ppm for all groundfish examined in the present study falls midway between North Sea and Irish Sea data.

Acknowledgments

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TABLE 6. *Mercury concentrations in plankton and sediment, North Atlantic offshore waters—1971*

Sample Data				Mercury Content
Latitude	Longitude	Depth, fathom	Date	Average, ppm wet weight
Plankton				
42° 50′	64° 32′	50-100	4-27	<0.05
43° 33′	68° 44′	55-110	4-21	<0.05
42° 13′	69° 59′	55-130	11-4	<0.05
43° 05′	68° 40′	60-120	11-17	<0.05
41° 29′	66° 47′	28-62	10-22	<0.05
36° 03′	74° 52′	28-62	4-4	<0.05
36° 51′	75° 20′	10-32	4-3	<0.05
39° 19′	73° 51′	10-32	4-1	<0.05
37° 14′	74° 58′	10-32	10-12	<0.05
Sediment				
42° 50′	64° 32′	50-100	4-27	<0.05
43° 33′	68° 44′	55-110	4-21	<0.05
43° 12′	70° 05′	55-130	11-8	<0.05
43° 05′	68° 40′	60-120	11-17	<0.05
43° 14′	68° 42′	60-120	11-17	<0.05
36° 03′	74° 52′	28-62	4-4	<0.05
36° 51′	75° 26′	10-32	4-3	<0.05
39° 19′	73° 51′	10-32	4-1	<0.05
40° 19′	70° 29′	23-62	10-15	<0.05
37° 14′	74° 58′	10-32	10-12	<0.05

TABLE 7. *Mercury concentrations in invertebrates, North Atlantic offshore waters—1971*

	Sample Data					Mercury Content		
						Average, ppm wet weight		
Species	Latitude	Longitude	Depth, fathoms	Date	Length, cm	Muscle	Whole Animal	Liver
Lobster (*Homarus americanus*)	39° 21′	72° 15′	95-200	4-9	6-19	0.31 [1]		0.60
Pandallid shrimp (unclassified)	42° 04′	68° 44′	58-115	4-19	ND		0.09	
	42° 48′	70° 38′	15-62	4-17	ND		<0.05	
	41° 19′	61° 20′	10-35	3-23	ND		<0.05	
Scallops (*Placopecten magellanicus*)	38° 10′	74° 07′	28-62	10-6	5-7	<0.05		—
Squid (*Illex illecebrosus*)	40° 13′	71° 07′	58-105	3-11	ND		<0.05	
	39° 38′	72° 60′	28-62	10-4	ND		<0.05	
	40° 02′	71° 11′	95-200	10-8	18-22		<0.05	
	36° 19′	74° 48′	95-200	4-4	ND		0.06	

NOTE: ND = no data.
[1] Lobster muscle sample from tail only.

Baseline Concentrations of Polychlorinated Biphenyls and DDT in Lake Michigan Fish, 1971 [1]

Gilman D. Veith [2]

ABSTRACT

Responding to the recommendations of the Lake Michigan Interstate Pesticide Committee, the author aimed to establish baseline data on polychlorinated biphenyls (PCB's) and DDT in Lake Michigan fish in 1971. Because the past 2 years had witnessed unprecedented legislative action to protect food resources and other aquatic species near the top of the food chain from persistent hazardous chemicals, the author also attempted to gauge the impact of cooperative legislative action on the quality of large lakes.

Thirteen species of fish taken from 14 regions of Lake Michigan in the fall of 1971 were analyzed for PCB's and DDT analogs. Mean wet-weight concentrations of PCB's similar to Aroclor 1254 ranged from 2.7 ppm in rainbow smelt to 15 ppm in lake trout. Most trout and salmon longer than 12 inches contained PCB's at concentrations greater than the tolerance level of 5 ppm established by the Food and Drug Administration, U.S. Department of Health, Education, and Welfare. Mean concentrations of total DDT ranged from less than 1 ppm in suckers to approximately 16 ppm in large lake trout. The presence of the major chlorinated hydrocarbons was confirmed by gas-liquid chromatography/mass spectrometry; additional PCB confirmations were obtained through perchlorination. The most abundant PCB's were tetra-, penta-, hexa-, and heptachlorobiphenyls which are similar to commercially prepared Aroclor 1254; lesser chlorinated PCB's were present in fish from nearshore waters.

[1] Supported by U.S. Environmental Protection Agency Project 16020 PBE, University Engineering Experiment Station, and University of Wisconsin Department of Civil and Environmental Engineering, Madison, Wis.

[2] National Water Quality Laboratory, U.S. Environmental Protection Agency, 6201 Congdon Boulevard, Duluth, Minn. 55804.

Introduction

This paper identifies and quantifies the most abundant organochlorine compounds, particularly polychlorinated biphenyls (PCB's) and DDT, in Lake Michigan fish in 1971. By establishing data on PCB's and DDT in Lake Michigan fish as recommended by the Lake Michigan Interstate Pesticide Committee, the author of the present study aimed to develop a 1971 baseline to predict trends of these chemicals in the lake. Lake Michigan contains much higher concentrations of potentially hazardous and persistent organic chemicals than the other Great Lakes, in part because of their widespread usage in the watershed and their disproportionally brief flushing period and low biomass density. Previous studies have shown that fish from Lake Michigan approach the action levels for dieldrin set by the Food and Drug Administration (FDA), U.S. Department of Health, Education, and Welfare (*1*); a major percentage of Lake Michigan fish exceeded the 5 ppm action level for DDT in 1969 (*2*). Similarly, Veith (*3*) has shown that PCB concentrations similar to Aroclor 1254 were greater than 15 ppm or three times the FDA action level in large fish captured from Lake Michigan in 1969.

Despite the comparatively high levels of DDT, dieldrin, and PCB's in Lake Michigan, there is no unequivocal evidence that they are endangering aquatic life. Concentrations of these chemicals appear to be below 10 parts per trillion (ppt) in the pelagic water and less than 100 ppt in nearshore waters. However, considerable indirect evidence suggests that the buildup of organochlorine compounds may threaten biological resources of the lake. Other reports have reviewed the chronic toxicity of pesticides and PCB's (*4-7*).

States surrounding Lake Michigan have made a major effort to restock the lake with brown, lake, and rainbow trout and coho and chinook salmon. Between 1963 and 1970, over 600,000 rainbow trout were released by Wisconsin alone (*8,9*). However, Reinert (*2*) previously noted that DDT and dieldrin levels in eggs of these fish are similar to concentrations which inhibited reproduction in the studies of Burdick et al. (*10*) and Macek (*11*). Johansson et al. (*12*) have shown that 15 ppm PCB's (lipid basis) in salmon eggs produced mortality in 50 percent of the samples tested. Death can be expected in all eggs when the PCB lipid content reaches 25 ppm.

Although continued stocking of fish may maintain a food resource, many fish contain residue body burdens which make them unfit for consumption. The effect of these fish on mink production in the North Central States has been studied in detail: before DDT and dieldrin concentrations in these fish had been well documented, Hartsough (*13*) indicated that the fish were suspected to inhibit mink reproduction; Aulerich et al. (*14*) clearly demonstrated that the fish had been the cause of the minks' reproductive failure; and Aulerich and Ringer (*15*) reported that DDT and DDD did not have significant adverse effects on mink. Furthermore, dieldrin was lethal to mink at 2.5 ppm in the food when fed for extended periods, but did not appear to affect reproduction at twice this concentration during the gestation period. Aulerich et al. concluded that feeding coho salmon to mink did not cause reproduction problems, but that the disorder is associated with other species of fish and ". . . appears to be dependent upon the species of fish and its environment" (*16*). Finally, after the earlier reports that PCB's were present in Lake Michigan fish, Ringer et al. (*17*) demonstrated that 10 ppm Aroclor 1254 in coho salmon produced 71 percent mortality in mink and that a mixture of 10 ppm PCB's and 0.5 ppm dieldrin in coho feed produced 100 percent mortality. No kits were born alive when the diet contained 5 ppm or more Aroclor 1254 alone. This clearly indicates that biological resources of Lake Michigan may seriously endanger other species even though concentrations of toxicants are not severe enough to produce readily discernible effects within the aquatic communities.

Equally important is the coincidence of high chlorinated hydrocarbon levels in herring gull and other bird populatons coupled with reproductive failures and subsequent population decline (*18*). Anderson (*19*) found that the eggs of the Great Lakes herring gull contained the highest chlorinated hydrocarbon levels ever reported for that species. He also found that the degree of eggshell thinning in the Lake Michigan gull, whose population declined dramatically in the early 1960's, varied from 9 percent in 1953-56 to 18 percent in 1965. In comparison, eggs of gulls on Lake Huron and Lake Superior have exhibited shell thinning of 7 percent and 8 percent, respectively, and those from gulls on the East Coast have remained essentially unchanged. Double-crested cormorants from Wisconsin had eggshells 20 percent thinner than those of gulls, and their eggs had the highest DDE concentrations of any cormorant eggs sampled from interior North America. Golden eagles, which feed primarily on mammals, do not show eggshell thinning as dramatic as that of bald eagles, which feed on fish (*19*).

Lake Michigan is the only Great Lakes watershed where major persistent chemicals have been curtailed. Although use of chlorinated pesticides in agriculture was probably diminishing in the late 1960's, the Lake Michigan Enforcement Conference recommended regulatory actions on many uses in 1968. This recommendation led to restrictions on DDT including its sale in Illinois, Michigan, and Wisconsin. A more detailed summary is presented by Lueschow (*20*). Monsanto Company, the sole producer of PCB's in the United States, restricted PCB sales in 1970; by April 1971 they were sold only to close-system users.

To measure the impact of the unprecedented cooperative legislative action regarding these chemicals, and to establish baseline data, the Lake Michigan Interstate Pesticide Committee recommended that this study be funded.

Sampling Procedures

Fish were collected in September and October 1971 with gill nets and pond nets from the four regions of Lake Michigan outlined in Figure 1. Whole fish were stored frozen (−20° C) in aluminum foil or polyethylene bags for 60 days or less and homogenized while frozen by repeatedly passing them through a meat grinder. All metal surfaces were rinsed with acetone, polyethylene bags were examined for interferences, and the grinder bearing and seal were checked periodically to assure that the sample was not contaminated during storage or preparation.

Analytical Procedures

REAGENTS

Sodium sulfate (Fisher Scientific Co.) was washed with three volumes of 1:1 hexane-acetone and dried at 130° C. To prevent further contamination from cap liners or containers, the Na_2SO_4 was stored in large glass bottles with aluminum foil liners in the cap.

The florisil (Kensington Chemical, Fisher Scientific Co.) was extracted in an all-glass Soxhlet extractor for 24 hours with the azeotrope of hexane and acetone to remove traces of organic impurities. The solvent was evaporated from the florisil at 100° C, and the solid was heated at 650° C for 2.5 hours for activation. If not used immediately after heating, the florisil was heated to 105° C before use.

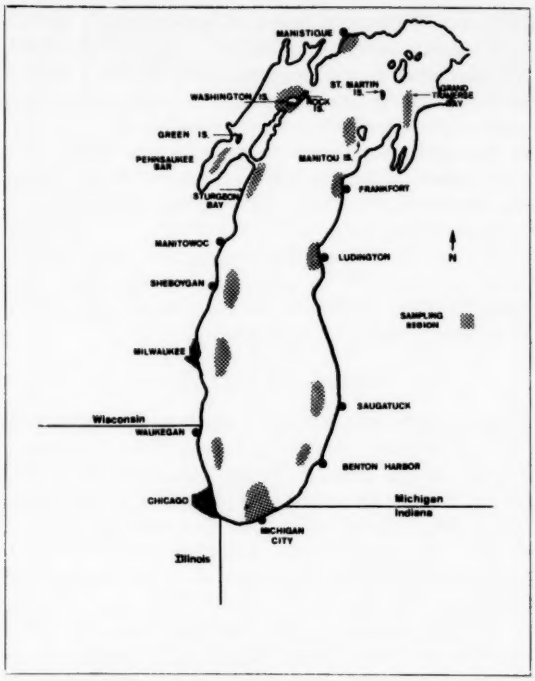

FIGURE 1. *Sampling regions for collecting fish, Lake Michigan—1971*

Other analytical components included:

Silicic acid:	Mallinckrodt Chemical, AR grade, Ramsey and Patterson. Used directly from reagent bottle.
Hexane:	Skelly B. Redistilled in glass from Dri-Sodium, Fisher Scientific Co.
Acetone:	Fisher Scientific Co., MCB. Redistilled in glass from Dri-Sodium, Fisher Scientific Co.
Ethyl ether and methylene chloride:	Mallinckrodt Chemical. Pesticide-quality solvents. Used directly from reagent bottle after periodic checks showed no interferences.
Glass wool:	Soaked in acetone, rinsed with 1:1 acetone:hexane mixture.
Glassware:	Washed thoroughly with hot detergent; rinsed once with hot water, twice with distilled water, again with 1:1 mixture of redistilled acetone:hexane.

PREPARATION OF SAMPLES

Procedures to extract and remove the bulk of the lipids have been described previously (*21*). Because of the high relative concentration of *p,p'*-DDE in Lake Michigan fish, DDE was quantitated directly by diluting 10 percent of the nonpolar florisil eluate to the appropriate volume for gas-liquid chromatographic (GLC) analysis. PCB's were separated from TDE and DDT isomers with a modified Armour and Burke procedure which omitted Celite 545 (*22*).

Quantitative gas chromatographic analyses were conducted on an Aerograph 1745-20 gas chromatograph equipped with dual concentric-tube electron-capture detectors (^{3}H, 250 mc). Columns were 2.0-m-by-1.8-mm-ID glass coils packed with 3 percent OV-101 on 120/140-mesh Gas-Chrom Q. The carrier gas, purified N_2, was maintained at 20 ml/min; the injector, column, and detector temperatures were 240°, 180°, and 220° C, respectively. Chromatograms were recorded on a Varian model A-25 dual pen recorder.

Previous work (*21*) showed that fish from Lake Michigan contain mixtures of PCB's that closely resemble the Aroclor 1254 produced by Monsanto Company, although PCB's both heavier and lighter than those most abundant in Aroclor 1254 were also present. The fish extracts contain predominantly those PCB's which elute at 70, 84, a doublet of 98 and 104, 125, 146, and 176; peak height of *p,p'*-DDE is represented here as 100 (Fig. 2). The presence of DDE precluded the use of the 98 and 104 PCB components in the quantitation, and PCB's based on Aroclor 1254 were determined by summing the heights of the 70, 84, 125, 146, and 174 PCB components when peak height of DDE is 100. This method also decreased the effect of minor compositional variations on the analytical result.

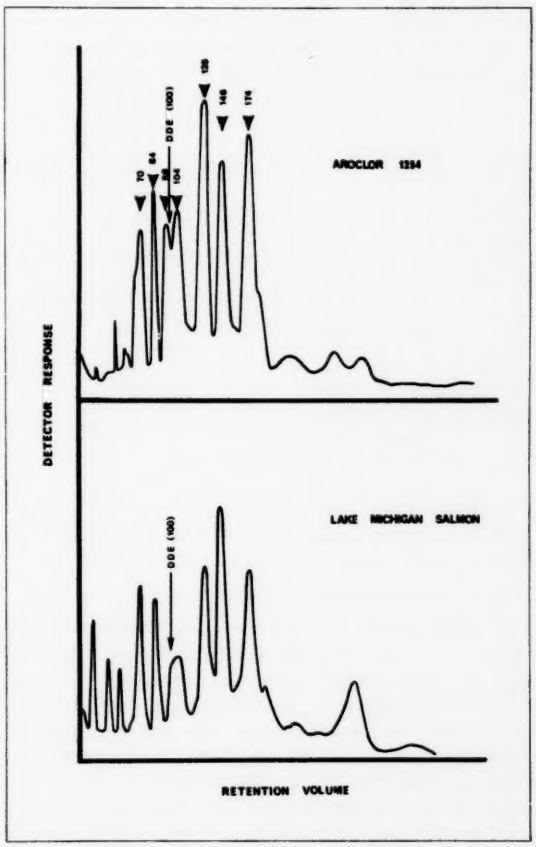

FIGURE 2. *Chromatograms of PCB mixtures in Aroclor 1254 and fish from Lake Michigan*

Recovery of PCB's from fish tissue averaged 85.1 ± 4.3 percent, whereas recovery of DDE was greater than 90 percent. The precision of the method outlined above is summarized in Table 1, which lists means and standard deviations of the analyses of six replicates of several fish species for PCB's, DDT, and lipids. The standard deviation for PCB analyses ranged from 5 percent in smaller fish to 14 percent in large coho salmon. The decrease in precision in analyses of large coho resulted from the difficulty of homogenizing larger fish. Precision was poorest in DDT analyses, where the standard deviation ranged from 8 to 23 percent with a mean of approximately 14 percent. This reduced precision results from losses during silicic acid chromatography which is used to quantitate the TDE and DDT isomers. The precision of DDE analyses was greater than those for DDT analyses, which was anticipated because of the fewer manipulations of the extracts. The standard deviation ranged from 7 to 18 percent, but the average deviation was approximately 10 percent. Because of the relatively simple procedure, lipid analyses were most precise, exhibiting standard deviations from 5 to 7 percent.

TABLE 1. *Precision of chlorinated hydrocarbon determinations for PCB's, DDE, DDT, and lipids in selected fish, Lake Michigan—1971*

Species	No. Replicates	Aroclor 1254	*p,p'*-DDE	*p,p'*-DDT	Lipid, %
Coho salmon (25 in.)	6	15.2±2.2	7.5±0.6	3.2±0.6	4.2±0.2
Coho salmon (27 in.)	6	13.1±1.3	5.5±0.4	1.8±0.4	10.0±0.6
Whitefish (13 in.)	6	4.1±0.2	0.35±0.04	0.56±0.04	16.2±0.9
Bloater (10 in.)	6	5.7±0.4	3.4±0.3	2.2±0.3	18.2±1.3
Alewife (8 in., composite)	6	4.0±0.2	1.1±0.2	1.0±0.2	5.7±0.3

NOTE: In columns 3-6, first number represents mean, second represents standard deviation.
Residues are ppm wet weight.

The determinable limit for PCB analysis was approximately 0.1 ppm; limits for *p,p'*-DDT, *o,p'*-DDT, *p,p'*-TDE, and *p,p'*-DDE were approximately 0.05 ppm.

CONFIRMATION OF MAJOR COMPONENTS

Major components of Lake Michigan fish extracts were characterized for a limited number of composite samples by standard gas-liquid chromatography/mass spectrometry techniques. In addition, the presence of PCB's in samples from each collection area was confirmed by perchlorination of PCB's to decachlorobiphenyl and subsequent analysis of the product by GLC (*23*). Aliquots of the hexane fraction of silicic acid columns were evaporated to dryness in a 5-ml vial fitted with a teflon-lined screw cap. Antimony pentachloride (0.2 ml) was added to the residue, and the vial was sealed and held at 180° C for 6 hours. Approximately 1 ml 6N MCl was added to the products to remove the $SbCl_5$ and the solution was extracted with five 1-ml hexane portions. The hexane was passed through a disposable pipette containing anhydrous Na_2SO_4 to remove traces of the aqueous solution. The sample was collected in a graduated centrifuge tube and diluted to the proper volume for GLC analysis. This technique also provided semiquantitative information for PCB's to supplement direct GLC analysis of the extracts. For those samples which contained PCB's similar to Aroclor 1254, estimates of total PCB's by perchlorination were within 15 percent of direct GLC analysis.

Results and Discussion

Approximately 850 fish were analyzed for PCB mixtures most closely resembling Aroclor 1254; a summary is presented in Table 2. Mean concentrations ranged from 2.7 ppm in smelt to 15.5 ppm in lake trout. Larger fish, such as brown, lake, and rainbow trout and chinook and coho salmon, contained PCB's at mean concentrations two to three times the 5 ppm action level established by FDA (*1*). Mean PCB concentrations in redhorse suckers, smelt, and whitefish were considerably less than 5 ppm. Mean concentrations in the alewife, carp, chub, and yellow perch were approximately 5 ppm; the range was 4.2-6.0 ppm. As expected, PCB levels increased with the percentage of fat and size of fish.

In Lake Michigan fish the mean concentration of total DDT, the sum of *p,p'*-DDT, *o,p'*-DDT, *p,p'*-TDE, and *p,p'*-DDE, ranged from 0.9 ppm in carp to 7.1 ppm in lake trout (Table 2). As with PCB's, fish with higher lipid concentrations contained greater concentrations of DDT.

The ratio of PCB's to total DDT ranged from 1.3 in redhorse suckers caught primarily in the northern waters to 7.6 in carp. The ratio of PCB's to DDT in the majority of the fish was between 1.7 and 2.8, and only in the carp, redhorse, yellow perch, and white sucker did the ratios fall outside this range. This ratio may become important in future studies to determine the rates at which the chemicals are eliminated from the Lake Michigan system.

Mean ratios of *o,p'*-DDT to *p,p'*-DDT ranged from 0.1 to 0.3; this ratio in the majority of the fish was 0.2. Since technical DDT generally contains about 30 percent of the *o,p'*-DDT isomer (ratio, 0.4), data from Lake Michigan fish suggest that degradation, other removal mechanisms, or both in the lake are slightly greater for the *o,p'* isomer than for the *p,p'* isomer. More than 80 percent of the total DDT residue is accounted for by *p,p'*-DDE and *p,p'*-DDT.

Not only did PCB concentrations vary considerably among the 13 species captured, but the range of PCB concentrations in a single lake species was generally

greater than 100 percent. The concentration range in red suckers was less, but all were captured in the same region of the lake. Although some variation in concentrations is expected because of the normal analytical error, the much larger ranges in Table 2 are undoubtedly due to other factors that limit usefulness of the mean concentrations presented. Previous research has shown that the lipid content, size of fish, season of capture, and concentration in the water may affect considerably the observed concentration of chlorinated hydrocarbons in tissue (*2*).

CHLOROBIPHENYLS

Regional variation in PCB concentrations and variations due to lipid content for each species are shown in Table 3. The wet-weight concentration of PCB's in ale-

TABLE 2. *Major chlorocarbons in fish, Lake Michigan—1971*

Species [1]	No. Fish Analyzed	Mean Fish Weight, g	Mean Lipid, %	Mean PCB's	Mean DDE	Mean ΣDDT	Mean PCB/ ΣDDT	Mean DDE/ ΣDDT	Mean *o,p'*-DDT/ *p,p'*-DDT
Alewife	85	100	6.5[3.9]	4.6[2.1]	1.7[0.8]	2.2[1.1]	2.4	0.8	0.2
Bloater	287	249	20.0[5.9]	6.0[2.2]	2.5[1.1]	3.8[2.8]	2.2	0.7	0.2
Brown trout	17	3,650	15.5[4.1]	7.3[2.8]	2.7[1.0]	4.2[1.6]	1.8	0.6	0.1
Carp	42	2,160	10.0[7.0]	4.2[3.6]	0.7[0.9]	0.9[1.2]	7.6	0.8	0.3
Chinook salmon	21	3,100	5.0[3.9]	11.4[4.0]	5.2[1.5]	6.8[2.5]	1.7	0.8	0.2
Coho salmon	56	2,720	6.5[2.1]	11.5[5.7]	4.8[2.3]	6.3[2.8]	2.1	0.7	0.2
Lake trout	134	1,620	16.6[4.3]	15.5[3.3]	5.0[2.8]	7.1[3.7]	2.5	0.7	0.2
Yellow perch	44	148	6.1[1.7]	5.8[3.5]	1.0[0.6]	1.6[1.1]	4.8	0.8	0.1
Rainbow trout	11	4,190	18.4[3.3]	9.3[4.1]	3.4[1.3]	4.2[1.8]	2.3	0.8	0.2
Redhorse sucker	16	902	8.6[1.2]	3.0[0.7]	1.6[0.5]	2.6[0.7]	1.3	0.6	0.2
Smelt	38	51	5.8[1.8]	2.7[1.3]	0.8[0.4]	1.2[0.6]	2.6	0.7	0.1
White sucker	51	1,130	5.9[2.8]	3.9[3.6]	1.0[0.5]	1.6[1.2]	3.4	0.7	0.1
Whitefish	43	1,170	17.6[4.4]	3.0[1.9]	0.8[0.3]	1.4[0.6]	2.8	0.7	0.2

NOTE: Expressions in brackets represent standard deviations.
Residues are ppm wet weight.
[1] Scientific names appear in Table 3.

TABLE 3. *Mean concentrations of PCB's and DDT in fish, Lake Michigan—1971*

Location	Capture Date	No. Fish Analyzed	PCB's	Percentage Fish Above 5 ppm PCB's	PCB's, Lipid Weight	*p,p'*-DDE	*p,p'*-TDE	*p,p'*-DDT	*o,p'*-DDT	Total DDT	Percentage Fish Above 5 ppm DDT
Alewife *(Alosa pseudoharengus)*											
Michigan City	10/15	10	4.4[2.0]	40	164	1.4	0.19	0.5	0.08	2.2	10
Benton Harbor	9/2	10	4.8[1.1]	40	60	1.8	0.23	0.5	0.08	2.7	0
Waukegan	8/23	2	2.5[0.1]	0	47	1.0	0.18	0.5	0.07	1.6	0
Saugatuck	4/11	12	5.3[2.2]	41	51	1.8	0.22	0.8	0.10	2.7	11
Sheboygan	10/15	4	5.5[1.4]	50	79	2.2	0.20	0.7	0.07	3.2	0
Ludington	7/3	16	4.4[1.4]	18	207	ND	ND	ND	ND	ND	ND
Frankfort	10/5	10	3.7[1.2]	20	62	1.3	0.16	0.4	0.06	1.9	0
Manitou Island	9/10	3	3.8[2.2]	33	51	3.2	0.33	1.3	0.16	5.0	30
Rock Island	9/11	3	8.9[12.0]	66	182	1.1	0.11	0.3	0.04	1.5	0
St. Martin Island	9/11	6	3.5[1.3]	16	82	1.0	0.14	0.3	0.05	1.5	0
Brown trout *(Salmo trutta)*											
Michigan City	10/15	1	11.9[0.0]	100	51	3.7	0.70	1.7	0.22	8.4	100
Sheboygan	7/13	5	7.9[3.0]	100	42	2.6	0.46	1.2	0.18	4.4	25
Gills Rock	9/16	10	6.7[2.5]	70	54	2.8	0.48	1.0	0.14	4.4	40
Carp *(Cyprinus carpio)*											
Michigan City	11/29	2	11.0[0.2]	100	72	3.3	0.81	0.3	0.14	4.6	50
Saugatuck	10/15	15	4.6[4.8]	26	71	0.6	0.20	0.1	0.02	0.9	0
Sheboygan	7/23	11	1.7[0.8]	0	30	0.2	0.07	0.0	0.01	0.3	0
Pensaukee Bar	11/1	9	4.2[1.4]	33	30	0.8	0.19	0.0	0.01	1.0	0
Manitou Island	9/16	5	5.8[0.8]	80	30	ND	ND	ND	ND	ND	ND
Bloater *(Coregonus hoyi)*											
Benton Harbor	9/2	10	5.0[1.2]	60	24	2.5	0.31	1.5	0.19	4.5	30
Saugatuck	6/16	18	8.1[1.9]	88	46	1.9	0.47	1.8	0.19	4.3	37
Saugatuck	6/18	24	7.8[2.2]	95	43	2.4	0.63	2.4	0.25	5.7	64
Saugatuck	6/19	15	6.9[1.8]	86	34	1.8	0.47	2.5	0.31	5.1	66
Milwaukee	9/19	11	4.6[1.2]	36	28	3.4	0.36	2.0	0.26	6.0	88
Sheboygan	7/13	13	5.1[1.5]	61	31	1.3	0.40	2.2	0.24	4.1	50
Sheboygan	7/22	13	6.1[1.8]	76	21	3.4	0.34	2.0	0.24	5.9	81

(Continued next page)

TABLE 3 (cont'd.). *Mean concentrations of PCB's and DDT in fish, Lake Michigan—1971*

Location	Capture Date	No. Fish Analyzed	PCB's	Percentage Fish Above 5 ppm PCB's	PCB's, lipid weight	*p,p'*-DDE	*p,p'*-TDE	*p,p'*-DDT	*o,p'*-DDT	Total DDT	Percentage Fish Above 5 ppm DDT
Sheboygan	8/19	6	3.8[0.6]	0	15	2.6	0.35	1.3	0.16	4.4	20
Sheboygan	8/27	7	5.0[1.3]	57	23	3.2	0.47	2.3	0.28	6.2	80
Sheboygan	10/15	6	3.7[0.8]	0	21	2.4	0.28	1.7	0.17	4.5	42
Ludington	7/3	43	7.4[1.8]	93	41	2.0	0.58	2.2	0.28	5.0	46
Frankfort	10/5	8	4.7[0.7]	12	28	2.2	0.33	1.2	0.16	3.8	0
Manitou Island	9/10	10	4.6[1.1]	30	24	3.1	0.50	1.8	0.26	5.6	50
Washington Island	9/9	6	4.1[0.6]	0	16	3.2	0.34	1.9	0.18	5.6	60
Rock Island	9/11	6	5.8[2.5]	66	97	2.8	0.36	1.1	0.08	4.4	20
Rock Island	9/14	7	5.6[1.6]	57	23	2.7	0.42	2.0	0.22	5.4	50
Rock Island	9/16	8	4.0[0.7]	12	18	2.8	0.29	1.3	0.14	4.5	40
St. Martin Island	7/19	9	3.4[0.6]	0	16	2.2	0.34	1.1	0.17	3.8	0
Manistique	5/27	9	4.8[0.7]	33	26	2.9	0.42	1.8	0.14	5.2	80
Chinook salmon (*Oncorhynchus tschawytscha*)											
Milwaukee	10/15	1	24.0[0.0]	100	117	6.6	1.67	4.7	0.53	13.6	100
Manitowoc	10/21	8	11.3[3.1]	100	209	6.3	0.62	1.2	0.21	8.3	100
Strawberry Creek	10/21	10	9.9[2.8]	100	373	4.5	0.39	1.0	0.17	8.0	88
Gills Rock	9/16	2	12.7[0.7]	100	278	5.5	0.44	1.2	0.20	7.3	100
Coho salmon (*Oncorhynchus kisutch*)											
Michigan City	4/17	8	3.6[1.7]	12	51	0.8	0.17	0.6	0.08	1.6	0
Michigan City	9/27	8	17.3[8.4]	87	255	5.3	0.46	1.2	0.20	7.2	85
Michigan City	10/7	6	14.0[4.0]	100	349	4.5	0.41	1.4	0.18	6.5	60
Sheboygan	10/21	9	12.1[2.8]	100	276	6.6	0.57	1.4	0.27	8.8	100
Ludington	8/28	4	11.2[3.3]	100	108	6.8	0.39	0.9	0.19	8.3	100
Platte River	10/7	10	12.9[1.3]	100	226	5.5	0.48	1.4	0.24	7.6	100
Gills Rock	9/16	11	12.6[4.1]	90	166	4.9	0.45	1.2	0.20	6.7	87
Lake trout (*Salvelinus namaycush*)											
Michigan City	10/15	4	14.9[2.0]	100	89	2.9	0.88	3.1	0.35	7.3	100
Michigan City	9/8	3	21.2[6.8]	100	121	9.5	1.12	3.9	0.45	14.9	100
Saugatuck	6/21	14	11.9[3.8]	100	70	4.7	0.65	2.1	0.27	7.6	88
Saugatuck	7/10	7	15.5[2.4]	100	78	6.9	0.70	2.0	0.24	9.8	100
Saugatuck	10/1	9	18.7[4.7]	100	113	6.7	0.84	2.8	0.27	10.6	100
Milwaukee	9/14	1	21.2[0.0]	100	90	10.1	0.96	3.0	0.47	14.5	100
Milwaukee	9/19	7	21.1[6.0]	100	114	10.6	1.05	3.3	0.45	15.4	100
Milwaukee	10/15	12	10.4[3.6]	100	127	2.3	0.44	1.3	0.15	4.2	20
Sheboygan	7/1	1	14.9[0.0]	100	58	ND	ND	ND	ND	ND	ND
Sheboygan	7/13	36	12.5[3.2]	100	82	4.3	0.51	1.7	0.24	6.8	64
Ludington	7/1	10	8.1[1.8]	100	51	3.3	0.52	1.6	0.22	5.6	60
Ludington	7/4	8	8.5[1.7]	100	58	3.2	0.63	2.0	0.30	6.1	66
Grand Traverse Bay	10/19	8	11.3[5.3]	75	56	6.0	0.73	2.0	0.30	9.0	77
Green Island	7/21	19	9.0[1.7]	100	48	3.8	0.50	1.5	0.21	6.0	72
Gills Rock	9/16	10	14.7[6.4]	100	83	5.5	0.62	1.9	0.24	8.3	62
Yellow perch (*Perca flavescens*)											
Michigan City	10/15	14	4.2[0.7]	14	72	0.8	0.15	0.4	0.04	1.3	0
Waukegan	8/28	9	6.1[1.5]	88	78	ND	ND	ND	ND	ND	ND
Milwaukee	10/13	10	10.9[3.1]	90	203	1.1	0.30	0.7	0.08	2.1	0
Ludington	7/6	3	6.2[1.3]	100	139	2.2	0.30	0.9	0.13	3.6	0
Frankfort	10/12	2	5.4[1.8]	50	69	1.7	0.43	2.2	0.14	4.5	0
Pensaukee Bar	8/25	10	2.7[2.1]	10	49	0.4	0.10	0.0	0.01	0.5	0
Rainbow trout (*Salmo gairdneri*)											
Michigan City	10/15	1	12.0[0.0]	100	66	ND	ND	ND	ND	ND	ND
Gills Rock	9/16	9	8.8[4.3]	77	47	3.1	0.34	0.9	0.16	4.5	40
Redhorse (*Moxostoma* sp.)											
Rock Island	9/11	7	2.8[0.9]	0	33	1.7	0.30	0.6	0.09	2.7	0
St. Martin Island	9/11	9	3.2[0.5]	0	37	1.5	0.35	0.8	0.09	2.7	0
Rainbow smelt (*Osmerus eperlanus modrax*)											
Michigan City	10/15	15	3.2[0.9]	0	45	0.9	0.11	0.4	0.04	1.4	0
Sheboygan	10/15	3	0.7[0.2]	0	18	0.6	0.12	0.3	0.03	1.0	0
Green Island	7/21	5	2.6[1.1]	0	57	0.7	0.12	0.2	0.02	1.0	0
Rock Island	9/11	8	2.9[1.7]	25	49	1.0	0.14	0.5	0.05	1.6	0
St. Martin Island	7/19	4	1.3[0.3]	0	31	0.7	0.09	0.2	0.04	1.0	0
White sucker (*Catostomus commersoni*)											
Michigan City	10/15	6	10.6[4.0]	100	100	1.8	0.32	1.8	0.08	4.6	66
Saugatuck	10/15	3	6.0[2.9]	66	149	0.7	0.14	0.2	0.03	1.0	0
Saugatuck	10/18	5	2.3[0.4]	0	40	ND	ND	ND	ND	ND	ND
Pensaukee Bar	8/6	7	3.2[3.1]	14	57	0.3	0.15	0.2	0.04	0.7	0
Grand Traverse Bay	10/19	14	2.3[0.7]	0	68	0.4	0.31	0.4	0.03	2.1	0
Green Island	7/21	1	2.1[0.0]	0	20	0.7	0.19	0.4	0.08	1.3	0
Rock Island	9/11	14	2.5[0.9]	0	46	0.9	0.20	0.3	0.04	1.4	0

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TABLE 3 (cont'd.). *Mean concentrations of PCB's and DDT in fish, Lake Michigan—1971*

Location	Capture Date	No. Fish Analyzed	PCB's	Percentage Fish Above 5 ppm PCB's	PCB's, Lipid Weight	*p,p'*-DDE	*p,p'*-TDE	*p,p'*-DDT	*o,p'*-DDT	Total DDT	Percentage Fish Above 5 ppm DDT
Lake Whitefish (*Coregonus clupeaformis*)											
Michigan City	10/15	4	6.1[0.9]	100	25	0.5	0.32	0.5	0.06	1.4	0
Saugatuck	6/19	7	2.7[0.5]	0	15	0.6	0.21	0.4	0.08	1.3	0
Saugatuck	6/21	2	3.1[1.3]	0	18	ND	ND	ND	ND	ND	ND
Grand Haven	9/6	7	5.8[0.9]	71	25	0.7	0.38	0.8	0.10	1.9	0
Grand Traverse Bay	10/19	13	1.8[0.3]	0	13	1.2	0.25	0.4	0.08	2.0	0
Rock Island	9/11	10	1.5[0.2]	0	9	0.7	0.16	0.4	0.05	1.4	0

NOTE: Expressions in brackets represent standard deviations.
ND = not determined.
Residues are ppm wet weight.

wives was greater in southern Lake Michigan than in the northern regions, although anomalies are apparent. Most alewives captured south of a line between Saugatuck and Sheboygan contained between 4.4 and 5.5 ppm PCB's, whereas those caught north of the line contained between 3.5 and 4.4 ppm. An interesting exception occurred in alewives from Rock Island just off the Door County Peninsula in Wisconsin; mean PCB concentration was 8.9 ppm.

Analysis of brown trout suggested similar trends: those from Michigan City at the southern end of the lake contained 11.9 ppm PCB's, whereas those from Sheboygan and Gills Rock contained 7.9 and 6.7 ppm PCB's, respectively.

Carp from Michigan City also had higher levels of PCB's than had those from the northern region. In contrast to the 11.0 ppm found in the Michigan City carp, those from Saugatuck and Sheboygan contained 4.6 and 1.7 ppm, respectively.

PCB's in bloaters in southern Lake Michigan ranged from 4.6 ppm near Milwaukee to 8.1 ppm near Saugatuck. In general, bloaters from the northern regions had concentrations below 5 ppm. The concentration of PCB's in the five groups of bloaters collected near Sheboygan during a 3-month period varied from 3.7 to 6.1 ppm, although the mean was below 5 ppm; no trend was indicated. The variation is somewhat less when data are expressed on a lipid basis; for example, PCB concentrations in the August 27 and October 15 bloaters were 5.0 and 3.7 ppm wet weight, respectively. In contrast, the concentration of PCB's in lipids was 23 ppm and 21 ppm, respectively. Thus much of the observed variation is caused by the variation in lipid content of fish.

All chinook salmon captured in Wisconsin contained more than 5 ppm PCB's; mean concentrations ranged from 9.9 ppm in Strawberry Creek (Sturgeon Bay) to 24 ppm at Milwaukee.

In concentrations of PCB's among coho salmon, authors observed little evidence of a trend dependent upon sampling region. Except for coho caught early in 1971 near Michigan City, mean concentrations of PCB's ranged from 11.2 ppm in salmon near Ludington to 17.3 ppm in those near Michigan City.

Among lake trout PCB concentrations were greatest in those from Michigan City, Saugatuck, and Milwaukee, where mean concentrations were generally between 15 and 21 ppm. Trout from the northern areas such as Sheboygan, Ludington, Grand Traverse Bay, and Gills Rock contained considerably less, and mean concentrations ranged between 8 and 15 ppm.

Mean concentration of PCB's was unexpectedly high, 10.9 ppm, in the 10 yellow perch caught near Milwaukee. Perch from other regions averaged less by a factor of two, and those from lower Green Bay contained 2.7 ppm.

In seven Rock Island redhorse, PCB residues averaged 2.8 ppm. In nine specimens from St. Martin Island, mean concentration was 3.2 ppm.

Rainbow trout were caught only near Michigan City and Gills Rock. The Michigan City rainbow trout had 12.0 ppm PCB's, whereas those from Gills Rock averaged only 8.8 ppm.

Concentrations of PCB's in white suckers and smelt were generally between 2 and 4 ppm, although the average concentration was 6.0 ppm in the three white suckers from Saugatuck on October 15 and 10.6 ppm in the six from Michigan City the same day. Except for whitefish caught at Grand Haven and Michigan City, the PCB concentration in whitefish was less than 3.1 ppm.

DDT AND ANALOGS

Concentrations of *p,p'*-DDT, *o,p'*-DDT, *p,p'*-TDE, and *p,p'*-DDE are presented in Table 3 along with the total DDT and percentage of fish that contained residues above the 5ppm action level established by FDA (*1*). Total DDT in alewives ranged from 1.6 ppm near Waukegan to 5.0 ppm near the Manitou Islands. There was no trend in the variations according to region. A brown trout from Michigan City contained 8.4 ppm total DDT, whereas those from Sheboygan and Gills Rock averaged 4.4 ppm. Except for the carp from Michigan City, which averaged 4.6 ppm total DDT, average concentrations in Lake Michigan carp were 1.0 ppm or less.

Total DDT in bloaters ranged from 3.8 ppm at Frankfort and St. Martin Island to 6.2 ppm at Sheboygan. There are no trends for DDT in chubs (Table 3). Except for eight coho salmon caught near Michigan City in the spring, which averaged 1.6 ppm total DDT, total DDT in this species varied little throughout the lake. Mean concentrations ranged between 6.5 and 8.8 ppm.

Lake trout from Michigan City averaged 14.9 ppm total DDT on September 8, 1971; those caught October 15, 1971, averaged only 7.3 ppm. The discrepancy is likely due to size differences. For example, the lake trout caught near Milwaukee in September also contained a mean concentration of approximately 15 ppm; however, the smaller trout caught near Milwaukee in October averaged only 4.2 ppm total DDT. DDT concentrations in lake trout from northern areas of the lake were less than 8 ppm; approximately 60-70 percent of the lake trout contained over 5 ppm total DDT.

Yellow perch from Pensaukee Bar in lower Green Bay had the lowest DDT content, 0.5 ppm; perch from other areas contained between 1.3 and 4.5 ppm total DDT. None of the perch contained more than 5 ppm total DDT.

Concentration of DDT in smelt, whitefish, and white suckers averaged approximately 1-2 ppm, although white suckers from Michigan City averaged 4.6 ppm total DDT.

Summary

Concentration of PCB's ranged from less than 2 ppm in small fish with low lipid content to over 20 ppm in larger fish with higher lipid content. The concentration of PCB's in Lake Michigan coho salmon is two to three times greater than in coho from Lake Huron, approximately 1.5 times greater than in Lake Ontario coho salmon, and approximately 10 times greater than in coho from Lakes Erie and Superior. Essentially 100 percent of large salmon and trout, both popular food sources, and 50-80 percent of bloaters from Lake Michigan contain PCB concentrations greater than the 5 ppm tolerance level set by FDA. Additional monitoring of this watershed is needed to determine whether tissue concentrations will reflect restrictions in domestic PCB sales and possible decreases in PCB usage in the watershed even though U.S. production of Aroclor 1254 has remained essentially the same as in 1969 (*24*).

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GENERAL

Distribution of Organochlorine Pesticides in an Agricultural Environment, Holland Marsh, Ontario—1970-72 [1]

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ABSTRACT

Analysis of organochlorine pesticides in soil, fish, and human blood samples from Holland Marsh, Ontario, indicates that although total DDT is present in detectable amounts, it does not constitute a hazard to human health and longevity. Among soils tested, residues were highest in surface samples. DDT levels in human blood samples were similar to those in U.S. and British studies.

Introduction

Holland Marsh, a 7,500-acre area devoted primarily to intense vegetable farming, is located 30 miles north of Toronto, Ontario. It is 7 miles long, 1-3 miles wide, and cultivated by 400 farmers. Soil is classified as peat muck and the average farm size is 25-30 acres. The marsh is served by eight cooperative packing houses which process 90 percent of the crop. Produce is then delivered to Toronto and Hamilton, Ontario; Detroit, Mich.; and areas of upper New York State including Buffalo and Rochester.

Aldrin, dieldrin, and DDT and its metabolites have been applied to soil and crops for the past 40 years. It is proposed that this survey of residues in the Holland Marsh ecosystem form a basis from which the ultimate fate of DDT residues in agricultural environments of southern Ontario may be determined. This study was commenced in 1970, 6 months after use of DDT was banned in Ontario.

Materials and Methods

Blood samples were taken from farm workers and packing house employees of both sexes. Ten-ml blood samples were collected in glass tubes containing potassium oxilate as an anticoagulant. Blood was collected in June (217 samples) and September (108 samples), the commencement and the end of the most active growing period.

Four farms situated along the north-south axis of the marsh were randomly selected for monitoring. Farm size and agricultural history are listed in Table 1. The four farms sampled were larger than average for the marsh, enhancing authors' opportunities for a large sampling area, personal interviews with each farmer, and accurate detailed history of the farm. Soil samples were collected during late spring from six sites randomly selected on each farm. Individual samples were taken from the soil surface, composite samples were taken from 0-7.5 cm deep, and five 7.5-cm cores were sampled in one location to a total depth of 45 cm on each farm. Sampling sites are shown in Figure 1.

TABLE 1. *History of farms sampled for DDT and related compounds, Southern Ontario—1970-72*

Farm	Acres Size,	Approximate Age, Year [1]	Soil Description	Crops Grown
1	135	20	Deep woody muck soil near canal but also sphagnum muck at north end. Very wet below 22.5 cm	Lettuce Celery Onions Carrots Potatoes Parsnips
2	180	28	Deep muck-peat soil. Little wood	Lettuce Carrots Celery Onions
3	30	30	Half is shallow muck with heavy clay underneath. Half is mineral soil with heavy clay underneath	Carrots Cauliflower Cauliflower Cabbage
4	100	10	Deep, wet muck soil. Very woody in places	Carrots Onions Lettuce

[1] Age of farm implies period of time land has been cultivated since first homesteaded.

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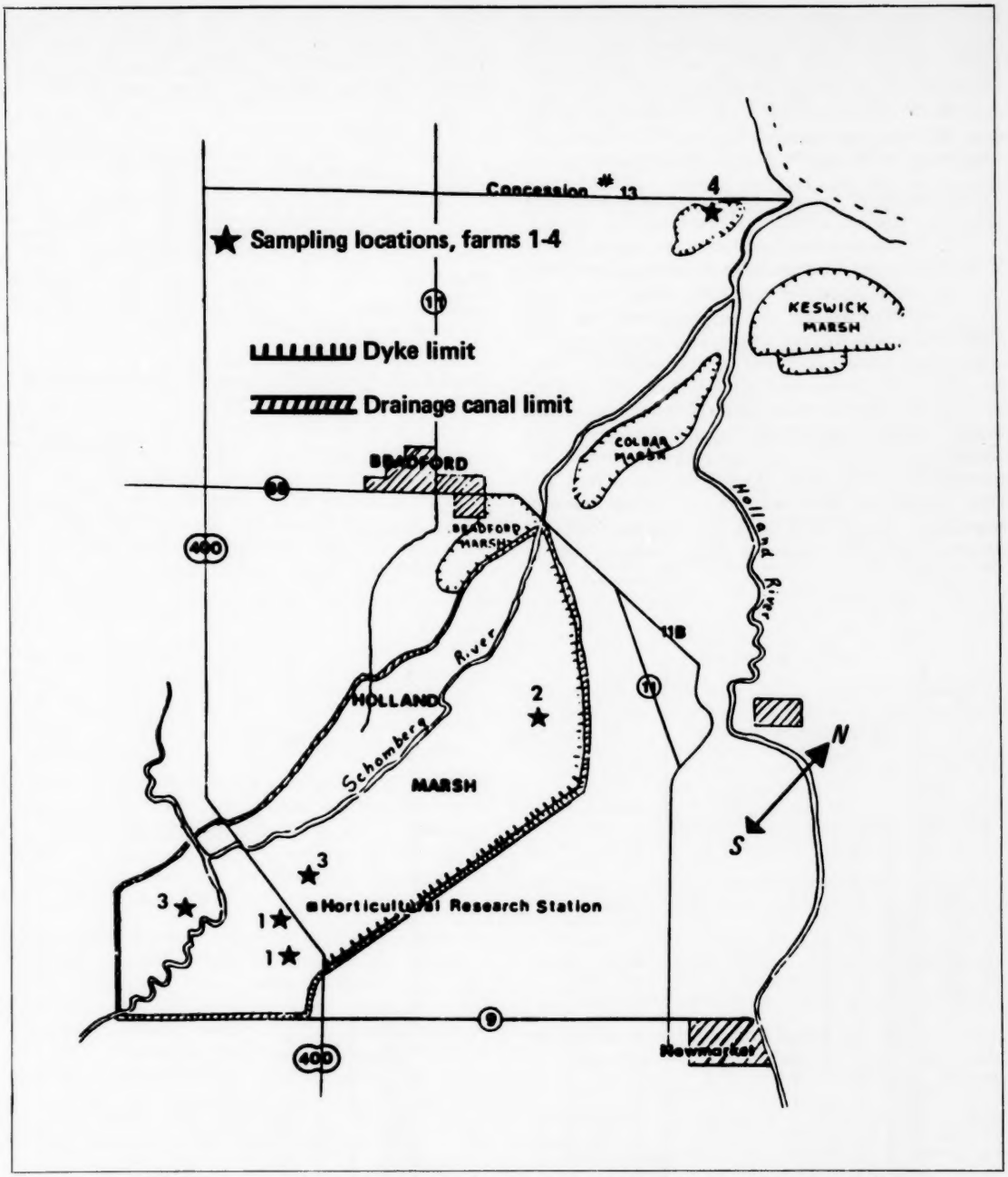

FIGURE 1. *Four farms in Holland Marsh, Ontario, sampled for DDT and related compounds*

Forty-eight fish were obtained by gill net or hook and line from streams throughout the marsh. Three ml whole blood was extracted with 10 ml acetonitrile using a Niagara shaker, 25 ml distilled water was added to the acetonitrile extract, pesticides were partitioned into hexane, and the hexane was concentrated to dryness with a rotary vacuum evaporator. The residue was redissolved in 2 ml hexane and used for subsequent gas-

liquid chromatographic (GLC) analysis. A Varian model 2100 gas chromatograph equipped with 250-mCi tritium electron-capture detectors was used. Columns measuring 1.8 by 4 mm ID were packed with 4 percent SE-30 + 6 percent QF-1 on 100-200-mesh chromosorb W. Average recovery rates for DDT, DDD, and DDE were 78, 82, and 79 percent, respectively.

Ten-g samples of fish and soil were mixed with 10 g anhydrous sodium sulfate and extracted by Soxhlet. Acetonitrile extracts were evaporated to approximately 5 ml, 50 ml sodium chloride solution was added, and the aqueous mixture was extracted three times with 50 ml redistilled hexane. The hexane extract was then evaporated to 10 ml and placed on top of a column containing a mixture of florisil and Celite in a 4:1 ratio by weight. The entire column was eluted with 200 ml of 6 percent ether in hexane and the eluate was evaporated to dryness with a rotary vacuum evaporator. The residue was redissolved in 4 ml hexane and used for subsequent GLC analysis. Average recovery rates for DDT, DDD, and DDE were 93, 93, and 91 percent, respectively. Results have been corrected for recovery. Sensitivity was 0.001 ppm for DDT, 0.0005 ppm for DDD, and 0.0001 ppm for DDE. Total DDT is calculated by adding DDT and the equivalent values for DDE (1.1148) and DDD (1.1076). Total DDT concentrations in human blood samples are listed in Table 2.

TABLE 2. *Total DDT in human blood samples, Holland Marsh, Ontario—1970-72*

	No. samples	Total DDT, ppm		
		Mean ± standard error	Median	Range, ppm
Whole group	356	0.016 ± 0.001	0.011	0.011-0.084
Farmers	92	0.019 ± 0.002	0.013	0.005-0.084
Packers and Others	264	0.014 ± 0.001	0.011	0.001-0.077

TABLE 3. *Hemoglobin in human blood, Holland Marsh, Ontario—1970-72*

	Sex	No. Samples	Mean ± Standard Error
Whole group	M	230	15.7 ± 0.1
	F	126	14.2 ± 0.2
Farmers	M	92	15.5 ± 0.2
Packers and others	M	138	16.1 ± 0.1
	F	126	13.8 ± 0.2

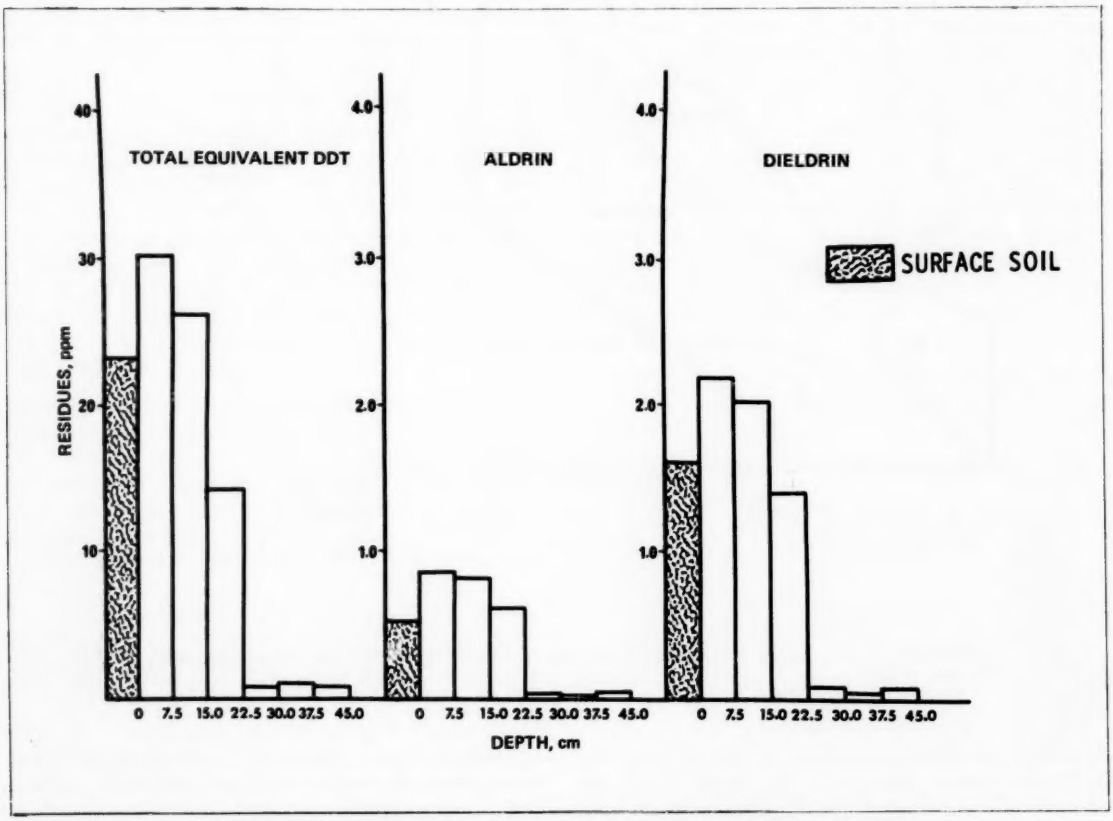

FIGURE 2. *Concentrations of organochlorine pesticide residues by soil depth*

TABLE 4. *Organochlorine pesticide residues in soil samples from four farms, Holland Marsh, Ontario—1970-72*

Depth, cm	No. Samples	Residues, ppm					
		Aldrin	Dieldrin	*p,p'*-DDE	DDD	*p,p'*-DDT	Total DDT
Surface	36	0.51	1.61	1.26	0.79	21.13	23.27
0- 7.5	18	0.86	2.19	1.64	1.02	27.41	30.38
7.5-15.0	18	0.78	2.03	1.49	1.07	23.49	26.38
15.0-22.5	18	0.62	1.40	0.86	0.85	12.35	14.25
22.5-30.0	18	0.03	0.08	0.07	0.02	0.73	0.83
30.0-37.5	18	0.02	0.06	0.08	0.03	0.85	0.97
37.5-45.0	18	0.04	0.08	0.08	0.03	0.82	0.95

Results and Discussion

Total DDT levels in human blood from Holland Marsh are similar to those given by Dale (*1*) for the general population in the United States (0.019 ppm) and by Robinson (*2*) for the general population in England (0.013 ppm). Marchand et al. (*3*) and Mastromatteo (*4*) have discussed the possibility of blood dyscrasias arising from the widespread use of chlorinated hydrocarbon pesticides. In view of these hypotheses, hemoglobin in all samples was analyzed using cyanomethemoglobin. Values derived (Table 3) were within clinically acceptable limits for both men and women and did not indicate blood dyscrasia.

Results of all human blood analyses were separated by month of sampling. Median values of total DDT from these two periods (0.013 and 0.011 ppm, respectively) were not significantly different. Quartiles around the medians are 0.010, 0.015, 0.010, and 0.012, respectively.

Samples from packing house employees had a median value of 0.011 ppm; those from the farmers had a median value of 0.013 ppm. As in the study by Wassermann et al. (*5*), people over 40 years of age had higher organochlorine pesticide residues than had those in younger age groups.

Mean residue levels of organochlorine soil samples are listed by farm and sample depth in Table 4 and illustrated in Figure 2. Most compounds detected are concentrated in the first 7.5 cm of soil, a distribution profile similar to that in soil of Norfolk County, Ontario (*6*). Similar results have also been obtained by Albright et al. in Alabama soils (*7*). Organochlorines are subject to decomposition by weathering and normal tillage. It is likely that total DDT concentrations in soil will diminish because DDT is no longer used in Ontario.

Fish samples had lower total DDT levels (Table 5) than those in similar species collected from other areas where DDT has been used extensively, such as Norfolk County, Ontario. Holland Marsh fish had higher DDT levels than fish taken recently from the base of the Bruce Peninsula, an unsettled area in Lake Huron.

The total DDT equivalent has been determined in various ecological systems in the Holland Marsh region. Results indicate that no serious health hazard is associated with total DDT concentrations present.

TABLE 5. *Total DDT in fish muscle, Holland Marsh, Ontario—1970-72*

Species		No. Samples	Mean Total DDT, ppm
Common Name	Scientific Name		
Brown bullhead	*Ictalurus nebulosus*	13	0.56
Bowfin	*Amia calva*	1	0.18
Carp	*Cyprinus carpio*	5	0.75
Goldfish	*Carassius auratus*	1	0.38
Golden shiner	*Notemigonus crysoleucas*	7	0.54
Yellow perch	*Perca flavescens*	5	0.39
Northern pike	*Esox lucius*	3	0.27
Rock bass	*Ambloplites rupestris*	2	0.71
Smallmouth bass	*Micropterus dolomieui*	1	0.73
White sucker	*Catostomus commersoni*	5	0.53
Sunfish (Pumpkinseed)	*Lepomis gibbosus*	15	0.46

NOTE: Fish were collected from ponds, streams, and rivers throughout the marsh.

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Organochlorine Pesticide Residues in a Farming Area, Nova Scotia—1972-73 [1,2]

B. G. Burns, M. E. Peach, and D. A. Stiles

ABSTRACT

Soil, silt, and water samples from the Habitant Creek watershed, Nova Scotia, a tobacco-growing area, have been monitored for organochlorine insecticides. Most samples contain measurable quantities of many persistent pesticides used in farming during the past decade. Sediment levels indicate that residues settle in sluggish parts of the stream. Drainage ditches show highest residual content caused in part by mass transport of soil in runoff. Residue content of water samples is normally one-tenth to one-hundredth that of silt, but is much higher during periods of heavy runoff. Levels vary with the seasons and are highest in the fall, decrease through the spring and summer, and are lowest in the winter. Although samples of well water taken fairly close to the stream showed virtually no residual content, a natural drainage reservoir had a pesticide content similar to that in the stream.

Introduction

In recent years the use of organochlorine insecticides in Canada has come under increasing scrutiny because of their long-term persistence. Although their role as general agricultural insecticides has diminished over the past 5 years, they were, until the early 1970's, used extensively to control infestation in tobacco.

Several reports (*1-5*) have appeared on the long-term persistence of organochlorine insecticides in a variety of settings, most of which were experimental plots. Although Harris (*2,6,7*) has extensively studied organochlorine residues in natural agricultural settings in southwestern Ontario, such work has generally received little attention.

During the past few years, the chemistry department at Acadia University has been investigating organochlorine pesticide residues in the Habitant Creek watershed of Nova Scotia. This region, approximately 10 square miles, is located in the Annapolis Valley and supports a variety of agriculture including tobacco farming. Soils in the agricultural part of the watershed are predominantly loam.

Sampling Procedures

Habitant Creek has both a tidal and a nontidal portion separated by an aboideau. This study deals with the nontidal section only and more specifically with the two main tributaries, Sleepy Hollow Brook and North Brook, which directly border farmland. Sampling locations are shown in Figure 1.

SILT

Samples were collected from readily accessible points along the banks and bed of the stream, natural drainage ditches, and other special areas such as the exit from a reservoir. Streambed samples were taken at a depth of 5-10 cm with a ladlelike device having a total volume of about 12 ml. Samples taken from stream banks and dry land were collected as soil cores of 2.5-cm diameter and 25-cm depth. Generally, at least three samples from the same site were thoroughly mixed prior to storage at 1.5° C.

WATER

Water samples were collected in 25-liter glass containers 1 m from the bottom of the stream. Because the waterway is relatively narrow (< 3 m) at all locations except site 30, no attempt was made to obtain vertical or horizontal profiles. However, it was felt necessary to ensure that all samples represented the stream as a whole rather

[1] Portions of this paper presented at the Third International Congress of Pesticide Chemistry, Helsinki, Finland, July 1974.

[2] Acadia University, Wolfville, Nova Scotia, Canada B0P 1X0. Study supported in part by Agriculture Canada Contract EMR 7103.

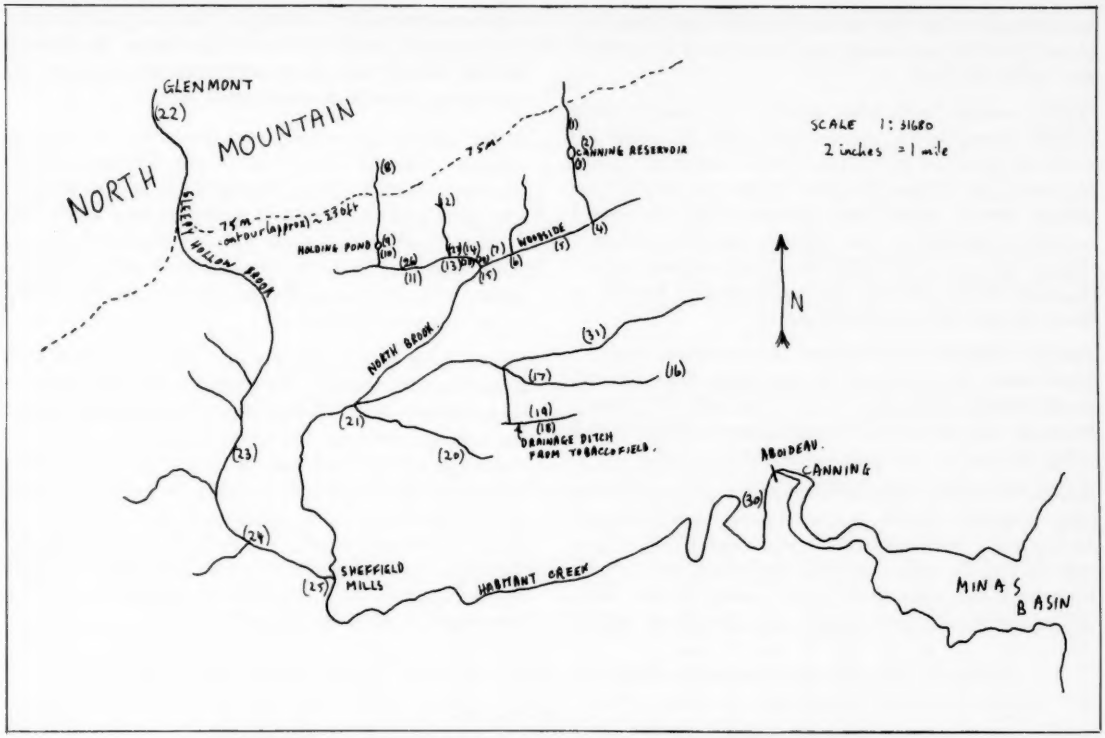

FIGURE 1. *Habitant Creek drainage system showing sampling locations*

than localized pockets. Consequently, three samples were collected at each location from positions as close as possible to the center of the stream. These were combined for analysis.

Analytical Procedures

Organochlorine insecticides were extracted from previously moistened silt samples according to the method of Peach et al. (*8*). Water samples were filtered through glass wool and extracted by the procedure of Kahn and Wayman (*9*). The combined water extracts were then cleaned in standard fashion (*8*).

Residues were identified by gas-liquid chromatography using the following operating parameters:

Gas chromatograph:	Hewlett-Packard F and M model 700
Column:	pyrex 1.22 m by 0.73 cm, packed with 3 percent OV-17 on acid-washed chromosorb-W, 60/80 mesh, or 1.83 m by 0.73 cm, packed with 3.8 percent UCW-98 on acid-washed chromosorb-W, 60/80 mesh
Detector:	electron-capture, pulsed at 50 μs
Temperature:	column 190° C detector 205° C
Purge/carrier gas:	95 percent argon / 5 percent methane Linde, dried by passing through molecular sieve
Flow rate:	purge gas 40 ml/min carrier gas 30 ml/min
Attenuation:	50 by 1

Peaks were identified by comparing their retention times with those of analytically pure standards applied to the above columns. Quantification was achieved by comparing peak areas. Calibration curves for each insecticide were prepared daily using analytical standards supplied by Montrose Chemical Corporation, Torrance, Calif.; Shell Canada Limited, Toronto, Ontario; and Velsicol Corporation, Chicago, Ill.

Percent recoveries ranged from 65 to just over 100 percent. Residue levels in soil and water samples were corrected for recovery.

Results and Discussion

The first series of samples was collected during the spring and early summer of 1972, mostly in the form of sediment from streambeds (Table 1). The sample from site 19 was taken from an uncultivated piece of land directly across the road from a drainage ditch serving tobacco fields. Most samples contained measurable quantities of commonly used organochlorine insecticides and their breakdown products.

Most frequently found were the two isomers of DDT in quantities which might be expected from a typical agricultural setting (*2*). Other residues were more localized

but difficult to link with particular field applications because these had been many and varied over a considerable period of time.

Highest residue levels were detected in a sample taken directly from site 18, the drainage ditch of a field on which tobacco had grown for 3 years. However, a sample taken just across the road at site 19, where little surface erosion could have occurred, had the lowest residues in the studied area. Samples were also collected during the summer of 1973 from places where natural drainage ditches led into the creek system. Results of these analyses are shown in Table 2.

Residue concentrations decrease in streambeds at points where water has moved slowly for some time. The exit to the holding pond (site 10) and the exit to Canning Reservoir (site 3) are the most obvious examples of sites where no residue was detected in 1972 or 1973.

Results also agree with water analyses conducted at two other drainage ditches where samples were collected during heavy prolonged rainshowers and about 3 days later (Table 3). Soil, especially the sandy loam of the Habitant Creek watershed, erodes during shower activity. Although the eroded soil was filtered off before water analyses were undertaken, the water still retained a substantial amount of insecticide. After the shower, the soil settled once more and aqueous pesticide concentrations reverted to more usual levels.

Water analyses have also been conducted on aqueous samples collected from several sites during different seasons. Although it was impossible to sample at some sites during the summer and winter because of too low a water flow or too much ice, overall results clearly show maximum pesticide concentration during fall and spring when rainfall is highest and the ground is most subject to erosion (Table 4).

Finally, samples of artesian water were taken from wells on properties bordering North Brook. Residue levels of water samples taken directly from the holding pond and the natural drainage reservoirs serving the towns of Canning and Wolfville are shown in Table 5. Artesian water contains virtually no residue, but wells depending on surface runoff have concentrations comparable to those of streams within the watershed. Although the Wolfville reservoir is not fed from the Habitant Creek system, analyses show that it has an organochlorine content similar to that of the creek.

TABLE 1. *Organochlorine insecticide residues in streambed sediments, Habitant Creek, Nova Scotia—1972*

Collection Site	Residues, ppb											
	γ-BHC	Heptachlor	Aldrin	Heptachlor Epoxide	Nonachlor	α-Chlordane	γ-Chlordane	Dieldrin	Endrin	*o,p'*-DDT	*p,p'*-DDT	Methoxychlor
1	7.9	0.0	5.3	0.0	6.2	42.2	2.5	3.0	5.1	0.9	154.0	0.0
2	11.6	0.0	42.0	0.0	5.0	38.0	0.0	6.0	5.6	22.0	274.0	0.0
3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
4	1.5	5.0	18.0	0.0	10.0	28.0	0.0	6.5	26.6	19.0	109.0	0.0
5	2.6	4.8	7.4	0.0	1.2	12.0	17.0	0.0	36.0	9.1	572.0	0.0
6	6.6	9.0	2.7	0.0	0.0	8.0	9.5	0.0	28.0	27.0	28.0	0.0
7	14.4	12.0	11.0	0.0	0.0	22.0	9.0	0.0	12.0	16.0	46.0	0.0
8	79.5	0.0	7.1	0.0	0.0	4.0	0.7	0.0	13.8	223.0	74.0	0.0
9	0.0	11.5	37.4	1.2	4.6	6.0	0.7	3.2	8.6	7.9	20.5	0.0
10	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
11	0.0	0.0	21.0	0.0	13.0	664.0	50.0	86.0	309.0	122.0	79.0	0.0
12	89.0	9.0	1.7	0.3	0.0	0.9	1.2	13.4	39.4	45.7	140.0	0.0
13	64.5	17.9	103.0	6.5	12.3	39.0	17.4	0.0	12.5	80.0	10.5	0.0
14	126.0	0.0	36.0	0.0	36.0	6.7	5.1	0.0	21.3	130.0	47.0	0.0
15	3.4	15.0	0.0	0.0	0.2	85.0	0.0	0.0	1.5	25.0	101.0	0.0
16	0.0	0.0	3.5	0.0	0.0	136.0	47.0	0.0	22.0	49.0	22.0	0.0
17	34.0	15.0	0.0	0.0	0.2	0.9	0.0	0.0	1.5	25.0	101.0	0.0
18	0.0	174.0	83.5	30.0	83.0	306.0	0.0	0.0	923.0	261.0	603.0	0.0
19	36.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
20	0.0	0.0	8.6	0.0	15.0	9.2	2.1	0.0	145.0	123.0	340.0	0.0
21	0.0	85.0	0.0	30.0	0.0	38.0	40.0	0.0	0.0	64.5	40.0	0.0
22	0.0	0.0	0.0	0.0	0.0	8.0	0.0	0.0	0.0	64.5	40.0	0.0
23	0.0	104.0	0.0	11.5	0.0	120.0	51.0	0.0	750.0	261.0	110.0	0.0
24	0.0	29.5	368.0	0.0	0.0	285.0	0.0	3.8	46.0	4.2	22.5	0.0
25	10.0	11.3	11.3	0.0	0.0	0.0	0.0	0.0	5.5	4.0	263.0	43.0

TABLE 2. *Organochlorine insecticide residues in sediments of natural drainage ditches entering Habitant Creek system, 1973*

Site	Residues, ppm											
	γ-BHC	Heptachlor	Aldrin	Heptachlor Epoxide	Nonachlor	α-Chlordane	γ-Chlordane	Dieldrin	Endrin	*o,p'*-DDT	*p,p'*-DDT	Methoxychlor
26	0.00	0.67	0.53	0.00	0.00	0.75	0.00	5.95	0.67	0.83	3.75	0.00
27	0.00	0.00	0.00	0.00	0.00	0.00	0.00	5.95	2.68	6.67	6.25	0.00
28	0.00	4.76	2.22	17.30	0.94	0.19	0.31	13.75	0.38	2.75	4.83	0.00
29	0.00	2.67	1.60	0.40	0.00	0.44	0.00	1.19	0.27	2.33	7.00	0.00

TABLE 3. *Organochlorine insecticide residues in drainage ditch water, Habitant Creek Watershed—1972*

Site	Residues, ppb											
	γ-BHC	Heptachlor	Aldrin	Heptachlor Epoxide	Nonachlor	α-Chlordane	γ-Chlordane	Dieldrin	Endrin	*o,p'*-DDT	*p,p'*-DDT	Methoxychlor
	During Thunderstorm											
18	0.08	0.38	0.33	1.04	0.47	0.46	0.42	0.00	1.07	0.49	0.45	0.05
Near site 6	0.06	0.00	0.02	0.00	0.00	0.45	0.04	0.00	0.31	0.56	0.64	0.02
	3 Days After Thunderstorm											
18	0.00	0.02	0.00	0.00	0.02	0.02	0.00	0.05	0.06	0.01	0.03	0.00
Near site 6	0.00	0.00	0.01	0.00	0.00	0.02	0.00	0.02	0.03	0.01	0.03	0.00

TABLE 4. *Organochlorine insecticide residues in water, Habitant Creek—1972*

Site, Date	Residues, ppb											
	γ-BHC	Heptachlor	Aldrin	Heptachlor Epoxide	Nonachlor	α-Chlordane	γ-Chlordane	Dieldrin	Endrin	*o,p'*-DDT	*p,p'*-DDT	Methoxychlor
22												
Spring	0.00	0.00	0.01	0.00	0.00	0.01	0.00	0.01	0.03	0.23	0.36	0.01
Summer	—	—	—	—	—	—	—	—	—	—	—	—
Fall	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.04	0.04	0.25	0.35	0.04
Winter	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
23												
Spring	0.00	0.10	0.55	0.43	0.41	3.72	0.00	0.29	0.12	0.15	0.15	0.09
Summer	0.00	0.36	0.42	0.05	0.08	0.56	0.00	0.47	0.01	0.18	0.18	0.00
Fall	0.11	0.46	0.61	0.86	1.67	2.41	0.30	1.12	0.73	0.36	1.49	0.09
Winter	0.01	0.00	0.00	0.00	0.02	0.05	0.00	0.00	0.01	0.05	0.08	0.00
24												
Spring	0.01	0.02	0.02	0.00	0.00	0.38	0.00	0.00	0.00	0.00	0.03	0.00
Summer	—	—	—	—	—	—	—	—	—	—	—	—
Fall	0.01	0.03	0.01	0.01	0.01	0.43	0.02	0.00	0.00	0.00	0.03	0.00
Winter	—	—	—	—	—	—	—	—	—	—	—	—
25												
Spring	0.01	0.26	0.03	0.02	0.04	0.01	0.00	0.04	0.03	0.05	0.05	0.00
Summer	—	—	—	—	—	—	—	—	—	—	—	—
Fall	0.02	0.08	0.12	0.20	0.67	0.25	0.08	0.03	0.04	0.04	0.09	0.00
Winter	0.03	0.00	0.00	0.03	0.03	0.07	0.00	0.05	0.00	0.02	0.06	0.00
30												
Spring	0.00	0.09	0.16	0.16	0.03	0.04	0.00	0.03	0.02	2.32	0.36	0.00
Summer	0.06	0.05	0.04	0.02	0.03	0.12	0.02	0.01	0.03	0.04	0.12	0.00
Fall	0.03	0.42	0.66	5.14	7.83	16.94	4.13	11.80	4.61	1.40	0.39	0.00
Winter	—	—	—	—	—	—	—	—	—	—	—	—
31												
Spring	0.01	0.02	0.01	0.01	0.00	0.01	0.00	0.01	0.03	0.05	0.01	0.00
Summer	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.11	0.01	0.00	0.00
Fall	0.01	0.03	0.01	0.01	0.01	0.13	0.00	0.01	0.04	0.05	0.10	0.00
Winter	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
21												
Spring	0.00	0.02	0.01	0.00	0.02	0.02	0.00	0.05	0.06	0.01	0.03	0.00
Summer	0.00	0.00	0.00	0.00	0.01	0.01	0.00	0.03	0.05	0.01	0.02	0.00
Fall	0.00	0.35	0.67	6.10	6.10	31.30	17.89	10.48	18.46	28.38	1.68	0.00
Winter	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

NOTE: — = no sample taken.

TABLE 5. *Organochlorine insecticide residues in waters within and outside Habitant Creek watershed, 1972-73*

Site	Residues, ppb											
	γ-BHC	Heptachlor	Aldrin	Heptachlor Epoxide	Nonachlor	α-Chlordane	γ-Chlordane	Dieldrin	Endrin	*o,p'*-DDT	*p,p'*-DDT	Methoxychlor
Artesian Wells												
1	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.01	0.00
2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Holding Pond	0.04	0.08	0.03	0.01	0.05	0.41	0.00	0.06	0.04	0.01	0.28	0.06
Canning Reservoir	0.02	0.05	0.04	0.08	0.21	0.86	0.27	0.22	0.16	0.09	0.30	0.00
Wolfville Reservoir	0.07	0.07	0.04	0.05	0.08	0.23	0.08	0.20	0.21	0.45	0.37	0.00

NOTE: Only the holding pond and Canning Reservoir are fed from Habitant Creek system.

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Chlorinated Hydrocarbon Pesticides and Mercury in Coastal Biota, Puerto Rico and the U.S. Virgin Islands—1972-74 [1]

Robert J. Reimold [2]

ABSTRACT

Baseline levels of mercury and chlorinated hydrocarbons were determined for Caribbean coastal biota as part of the U.S. Environmental Protection Agency estuarine monitoring program. Forty-one percent of the 150 environmental samples taken had significant levels of these compounds.

Concentrations of chlorinated hydrocarbons suggest spatial and temporal variations within the plant or animal. In some cases residues in biota could be related to the land-use practices in the sampled watershed.

Introduction

The presence of chlorinated hydrocarbons in continental U.S. marine and estuarine organisms has been routinely monitored since 1965 (*1, 2*). There is, however, a paucity of data concerning concentrations of these chlorinated hydrocarbons in estuarine and marine fauna and flora from the Caribbean (*3*).

As part of the estuarine monitoring program of the U.S. Environmental Protection Agency (EPA), a biannual survey of selected Caribbean islands was initiated in October 1972. The purpose of this paper is to report baseline concentrations including negative results in selected environmental samples collected from U.S. territories in the Caribbean.

Methods

The study area includes Puerto Rico and the U.S. Virgin Islands of St. John, St. Thomas, and St. Croix. Collection locations for each of the four island areas are identified in Figures 1 and 2. At each location, samples were collected within 1 km of the edge of shore. Sites were selected in watersheds which respond quickly to rainfall and produce runoff which might contribute pollutants to coastal biota.

Samples were collected by seine, trap, hook and line, or by hand during fall 1972, spring 1973, fall 1973, and spring 1974. They were immediately placed on ice in aluminum foil packets. Within 4 hours of collection, samples were processed for analysis according to previously developed techniques (*4,5*). Operating parameters of the gas-liquid chromatographic (GLC) techniques were:

Column:	glass, 5 ft by 18 in., packed with 3 percent DC-200 on 80/100 mesh Gas-Chrom Q
Temperatures:	Oven 190° C Injector 210° C Detector 210° C
Carrier Gas:	Prepurified nitrogen at a flow rate of 40 ml/min. Three columns of different polarity were used for confirmation.

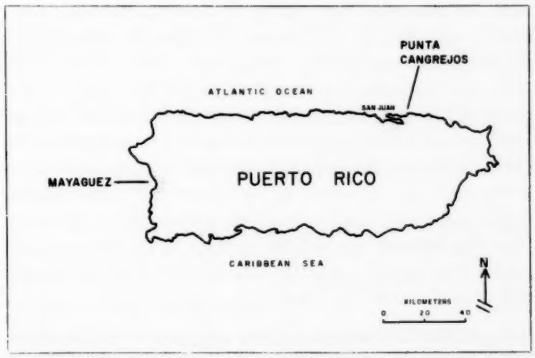

FIGURE 1. *Collection sites for coastal biota, Puerto Rico*

[1] Contribution No. 286, Marine Institute, University of Georgia, Sapelo Island, Ga. 31327. Project funded in part by U.S. EPA Contract 68-02-1254.

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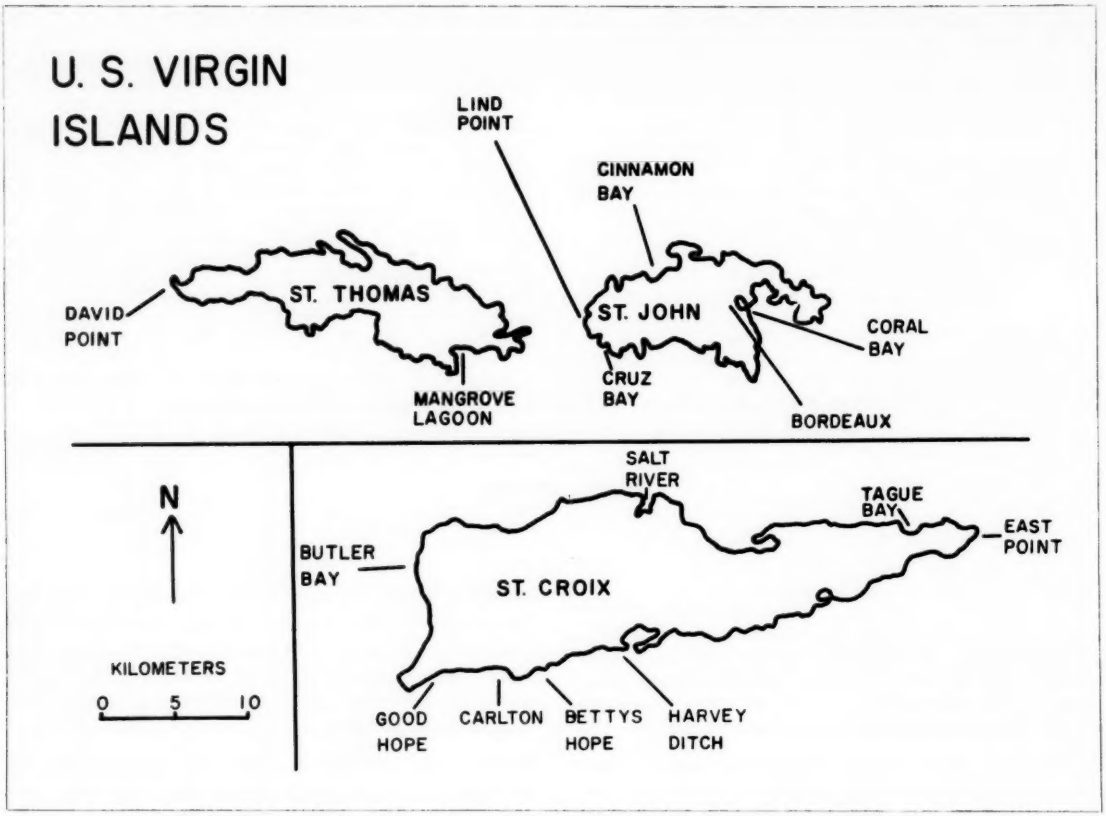

FIGURE 2. *Collection sites: St. Croix, St. Thomas, and St. John, U.S. Virgin Islands*

Analyses were performed by the EPA Pesticide Monitoring Laboratory, Bay St. Louis, Miss., and by the Marine Institute Laboratory, University of Georgia, Sapelo Island. In an earlier monitoring program for chlorinated hydrocarbon pesticides, the two laboratories synchronized methodologies and analyzed split samples. Results from this study were not significantly different.

Samples processed by the EPA laboratory were analyzed using Butler's technique (*2*). Specific chlorinated hydrocarbons determined were DDT, DDE, TDE, dieldrin, and polychlorinated biphenyls (PCB's). The PCB's were separated from pesticides and identified as Aroclor 1254 by matching chromatograms of field samples with chromatograms of standard Aroclor samples. All concentrations are reported as μg/kg or ppb on a whole-body, wet-weight basis. Relative recovery from the samples was between 85 and 90 percent. Data are not corrected for recovery. Concentrations less than 5 ppb are not considered in this study.

All analyses for mercury were conducted by the EPA laboratory using the techniques of Uthe et al. (*6*) and Brandenberger and Bader (*7,8*). These results are also expressed in μg/kg or ppb on a whole-body, wet-weight basis. Mercury concentrations less than 0.02 μg/kg are not considered.

Results

Of 150 environmental samples collected and analyzed, 41 percent had significant concentrations (> 5 ppb) of mercury or chlorinated hydrocarbons (Table 1). Table 2 lists scientific names of the biota and summarizes sample collection data.

Low concentrations of dieldrin were found in red mangrove leaves collected from St. Croix and St. John. There were also very low concentrations of dieldrin in a Nassau grouper and a soldier crab, both from St. John. Occurrence of dieldrin in St. John samples from the Coral Bay coincides with the presence of dieldrin in well and cistern water from a church at the bay (*9*). Cistern water sampled in 1970 contained 0.01 ppb dieldrin.

TABLE 1. *Chlorinated hydrocarbon concentrations in biota, U.S. Virgin Islands and Puerto Rico, 1972-74*

Date	Location	Common Name	Residues, ppb					
			Dieldrin	DDT	DDE	TDE	PCB's	Mercury
Fall '72	Mayaguez PR	Snook		157			162	
Spring '73	Mayaguez PR	Great barracuda		69	103			968
Spring '73	Mayaguez PR	Red drum						114
Fall '73	Mayaguez PR	Bluestriped grunt						70
Spring '74	Mayaguez PR	Yellowfin mojarra					399	
Spring '74	Mayaguez PR	Barbu					157	824
Fall '72	Punta Cangrejos PR	West Indian sardine		39		25	201	
Spring '73	Punta Cangrejos PR	West Indian sardine					416	271
Spring '73	Punta Cangrejos PR	Atlantic spadefish					240	194
Spring '73	Punta Cangrejos PR	Red mangrove						40
Spring '73	Punta Cangrejos PR	Great barracuda		69	103			968
Fall '73	Punta Cangrejos PR	Yellowfin mojarra						87
Spring '74	Punta Cangrejos PR	Striped mullet					579	
Spring '74	Punta Cangrejos PR	Silver jenny					89	
Spring '74	Punta Cangrejos PR	Red mangrove					68	80
Spring '74	Punta Cangrejos PR	Fiddler crab		<10	12	<10		
Fall '72	David Point STT	Red grouper					133	
Spring '73	David Point STT	Queen triggerfish						400
Spring '73	David Point STT	Red snapper						736
Fall '73	David Point STT	Queen triggerfish						822
Fall '73	David Point STT	Rock hind						290
Spring '74	David Point STT	Rock hind						304
Spring '74	David Point STT	Gray snapper						700
Spring '74	Mangrove Lagoon STT	Red mangrove					64	20
Fall '72	Cruz Bay STJ	Soldier crab		36	44	14		
Fall '72	Cruz Bay STJ	Red mangrove					129	
Fall '73	Cruz Bay STJ	Rock hind						40
Fall '72	Cinnamon Bay STJ	Mongoose (liver only)		22	47	11		
Fall '72	Coral Bay STJ	Soldier crab		43	132	14		
Fall '72	Coral Bay STJ	Red mangrove	21					
Fall '72	Coral Bay STJ	Turtle grass					40	
Fall '72	Coral Bay STJ	Nassau grouper	<10					
Spring '73	Coral Bay STJ	West Indian sardine					719	
Spring '73	Coral Bay STJ	Mangrove oysters						58
Spring '73	Coral Bay STJ	Schoolmaster						155
Spring '73	Coral Bay STJ	Porcupine fish						78
Fall '73	Coral Bay STJ	Red mangrove						50
Spring '74	Coral Bay STJ	Mangrove oysters					110	
Fall '73	Bordeaux STJ	Soldier crab	<10	30	112			
Fall '73	Bordeaux STJ	Soldier crab		758	17508			
Fall '73	Lind Point STJ	Soldier crab			21			
Fall '72	East End Point STX	Jewfish					495	
Fall '73	East End Point STX	Yellowtail snapper						83
Fall '73	East End Point STX	Ocean surgeon						25
Fall '73	East End Point STX	Queen triggerfish						75
Fall '72	Harvey Ditch STX	Red mangrove					181	
Fall '72	Harvey Ditch STX	Red mangrove					181	
Spring '73	Harvey Ditch STX	Red mangrove						38
Spring '74	Harvey Ditch STX	Striped mullet					809	
Spring '74	Harvey Ditch STX	French grunt					467	
Spring '74	Harvey Ditch STX	Spotted pink shrimp					4630	
Spring '74	Harvey Ditch STX	Fiddler crabs	<10				8624	
Spring '74	Harvey Ditch STX	Striped mullet					5391	
Fall '72	Bettys Hope STX	Red mangrove					132	
Fall '72	Bettys Hope STX	Checkered puffer					1817	
Fall '72	Carleton STX	Red mangrove					61	
Spring '73	Carleton STX	Red mangrove	11					
Fall '72	Good Hope STX	Fish doctor					166	
Fall '72	Butler Bay STX	Great barracuda					129	
Spring '73	Salt River STX	Mangrove oysters						39
Spring '73	Tague Bay STX	Great barracuda						271

NOTE: All residues are given on a whole-fish, wet-weight basis.
PR = Puerto Rico, STJ = St. John, STT = St. Thomas, STX = St. Croix.
Blank spaces indicate no residues detected.

DDT and DDE were found in mongoose liver collected from Cinnamon Bay, St. John, and soldier crabs collected at Coral Bay and nearby Bordeaux on St. John. Earlier studies of the cistern water and sediments at these sites did not reveal DDT or DDE (*9*), although they reported 0.14 ppm DDE in sediment of a cistern at Coral Bay.

DDT and DDE were found in East Indian sardines, snook, and great barracuda from Mayaquez, P.R. In the great barracuda, there were 69 μg/kg DDT and 103 μg/kg DDE on a whole-fish basis. Giam et al. (*3*) sampled great barracuda east of the Yucatan peninsula and detected 8 μg/kg DDT in the muscle and 42 μg/kg DDT in the liver.

TDE was found only in soldier crabs (14 μg/kg) and mongoose liver (11 μg/kg) from Cruz, Coral, and Cinnamon Bays, St. John. Earlier research efforts (*9*) did not reveal TDE at these sites.

Aroclor 1254, the only PCB compound identified, was found in West Indian sardines collected from Mayaguez,

P.R. (201 μg/kg), Punta Cangrejos, P.R. (416 μg/kg), and Coral Bay, St. John (719 μg/kg). This compound also appeared in red mangrove leaves from Cruz Bay, St. John (129 μg/kg); and from Harvey Ditch (181 μg/kg), Bettys Hope (132 μg/kg), and Carleton (61 μg/kg), all in St. Croix. Aroclor 1254 was also detected in several other fish (Table 1) including a great barracuda from Butler Bay, St. Croix (129 μg/kg), and striped mullet (5,391 μg/kg) and fiddler crabs (8,624 μg/kg) from Harvey Ditch, St. Croix. This is a considerably greater concentration of Aroclor 1254 than that reported in a great barracuda collected by Giam et al. (*3*), which had 9 μg/kg in the muscle and 57 μg/kg in the liver.

TABLE 2. *Genera, species, and common names of biota sampled, U.S. Virgin Islands and Puerto Rico, fall 1972—spring 1974*

SPECIES	PR	STJ	STT	STX
PLANTS				
Red mangrove (*Rhizophora mangle*)	X	X	X	X
Turtle grass (*Thalassia testudinum*)		X		
INVERTEBRATES				
Spotted pink shrimp (*Penaeus brasiliensis*)				X
Fiddler crab (*Uca pugilator*)	X			X
Soldier crab (*Coenobita clypeatus*)		X		
Pacific oysters[1] (*Crassostrea gigas*)				X
Mangrove oysters (*Crassostrea rhizophorae*)		X		X
Scallops[1] (*Argopecten irradians*)				X
FISH				
Ocean surgeon (*Acanthurus bahianus*)				X
Spotted eagle ray (*Aetobatus narinari*)				X
Queen triggerfish (*Balistes vetula*)			X	X
Bar jack (*Caranx ruber*)				X
Snook (*Centropomus undecimalis*)	X			
Atlantic spadefish (*Chaetodipterus faber*)	X			
Banded butterflyfish (*Chaetodon striatus*)				X
Porcupinefish (*Diodon hystrix*)		X		
Rock hind (*Epinephelus adscensionis*)	X	X	X	X
Jewfish (*Epinephelus itajara*)				X
Red grouper (*Epinephelus morio*)			X	
Nassau grouper (*Epinephelus striatus*)			X	X
Silver jenny (*Eucinostomus gula*)	X			
Yellowfin mojarra (*Gerres cinereus*)	X			X
Fish doctor (*Gymnelis viridis*)				X
Margate (*Haemulon album*)				X
French grunt (*Haemulon flavolineatum*)				X
White grunt (*Haemulon plumieri*)				X
Bluestriped grunt (*Haemulon sciurus*)	X	X		
Schoolmaster (*Lutjanus apodus*)	X	X		X
Red snapper (*Lutjanus campechanus*)			X	
Sand tilefish (*Malacanthus plumieri*)				X
Striped mullet (*Mugil cephalus*)	X			X
White mullet (*Mugil curema*)	X			
Yellowtail snapper (*Ocyurus chrysurus*)				X
Barbu (*Polydactylus virginicus*)	X			
Spotted goatfish (*Pseudupeneus maculatus*)		X		
West Indian sardine (*Sardinella sardina*)	X			
Striped parrotfish (*Scarus croicensis*)	X			
Red drum (*Sciaenops ocellata*)	X			
Redtail parrotfish (*Sparisoma chrysopterum*)				X
Stoplight parrotfish (*Sparisoma viride*)		X	X	X
Checkered puffer (*Sphoeroides testudineus*)				X
Great barracuda (*Sphyraena barracuda*)	X			X
MAMMALS				
Mongoose (*Herpestes javanicus*)				X

NOTE: PR = Puerto Rico, STT = St. Thomas, STJ = St. John, STX = St. Croix.

[1] Sample obtained from artificial upwelling mariculture project, Lamont-Doherty Geological Observatory, Columbia University.

The highest concentration of mercury, 968 μg/kg, was found in a great barracuda collected near Mayaguez, P.R., in spring 1973. It was also detected in the red snapper (400 μg/kg) collected at David Point, St. Thomas. Mercury also occurred in the red mangrove from Punta Cangrejos, P.R. (40 μg/kg) and from Harvey Ditch, St. Croix (38 μg/kg); and in other components low in the food chain, such as mangrove oysters from Coral Bay, St. John (58 μg/kg), and from Salt River, St. Croix (39 μg/kg).

Discussion

The author's findings supplement information recently published by Giam et al. (*3*) and Lenon et al. (*9*). In all three studies, concentrations of chlorinated hydrocarbons suggest spatial and temporal variation within the same plant or animal.

In some instances, residues in the biota could be related to the land-use practices in the watershed. At Coral Bay, St. John, the Virgin Islands Department of Health had employed both DDT and dieldrin in an insect control program. Lenon et al. (*9*) reported both DDT and dieldrin in cistern water at Coral Bay. As shown in Table 1 of the present study, dieldrin and DDT were found in measurable concentrations in fauna collected from Coral Bay.

In another instance illustrated in Table 1 and Figure 2, Aroclor 1254 was found in red mangroves on the south side of St. Croix. Residues in samples collected westward along the ocean shore during fall 1972 declined from the first measured concentration, 181 μg/kg at Harvey Ditch, to 132 μg/kg at Bettys Hope and 61 μg/kg at Carleton. By spring 1974, Aroclor 1254 had trophically magnified to levels of 8,624 μg/kg in the fiddler crabs and 5,391 μg/kg in striped mullet, both potential detrivores. The source of this PCB compound is not known. One might speculate that it is local, considering that residues are found at such low trophic levels in the primary producers and detrivores.

It is not yet possible to determine whether the detected measured pollutants are increasing, decreasing, or maintaining status quo. Data in this study do establish baseline conditions for future comparisons in these relatively uncontaminated areas of Puerto Rico and the U.S. Virgin Islands.

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Total Mercury in Water, Sediment, and Selected Aquatic Organisms, Carson River, Nevada—1972 [1]

Robert T. Richins [2] and Arthur C. Risser, Jr.[3]

ABSTRACT

A 1971-72 study of the Nevada Carson River drainage system by the Geological Survey, U.S. Department of Interior, revealed substantial amounts of mercury from pre-1900 gold and silver milling operations of the Comstock Lode. A monitoring survey was initiated to determine the extent of mercury uptake from corresponding surface water and sediments for seven aquatic species collected from five sampling stations along the watercourse. Total mercury content in fish ranged from 0.02 to 2.72 ppm; highest concentrations occurred in piscivorous white bass (0.50-2.72 ppm) sampled from Lahontan Reservoir. Residue levels appeared to be related to fish size, as demonstrated by highly significant correlations between wet weight and mercury content of five of the six species. Concentrations also appeared to be directly influenced by the species' position on the aquatic food chain. These results indicate that mercury levels in some fish from the Carson River drainage system may exceed the 0.50 ppm maximum concentration considered by the Food and Drug Administration, U.S. Department of Health, Education, and Welfare, to be safe for human consumption.

Introduction

Increased mercury levels in various localized areas, particularly the aquatic environment, are often influenced by people. Their use of mercury has threatened bird populations in Sweden (*1*) and contaminated lakes and rivers in some areas of the United States, rendering fish unsafe for consumption (*2*). Consequences of exposure to such acute concentrations are evident in reports from Minamata and Niigata, Japan (*3,4*).

Natural sources also contribute to mercury contamination (*5*). Levels of naturally occurring mercury as high as 1,200 ppb have been reported in air over heavy ore deposits (*6*). It has been estimated that the earth receives approximately 100,000 tons of mercury annually from precipitation. This compares to the yearly human production of about 10,000 tons (*7*).

Since the first century B.C. elemental mercury has been used to extract gold and silver from their ores by amalgamation (*8*). Thus when the Nevada Comstock Lode was discovered in the spring of 1859 near Virginia City, Nev. (*9*), large amounts of the liquid metal were imported to 75 gold-milling sites in the area. In 1869 railroad lines connected Virginia City with the 12 millsites along the Carson River in the Brunswick Canyon area. After this link was completed nearly all milling in the Comstock Lode was carried out at these sites because water power was available (*10*).

The Patio process, which employed an average charge of 1:10 quicksilver (mercury) to the weight of the ore (*9*), was used for extraction (*11*). Though records are incomplete, Hatch and Ott's estimates of total mercury lost during the 30-year peak of the Comstock (1865-1895) are as high as 200,000 flasks, or approximately 15,000,000 lb (*12*). These authors also make further reference to the recovery of quicksilver from tailings at the Douglas Mill in Six-Mile Canyon below Virginia City. Here, using cyanide and flotation methods which had first been perfected for extracting gold and silver, the site's tailings were refined around 1906. Between 1906 and 1914 the operation recovered over 1,000 flasks of mercury.

Until the last decade it was generally accepted that metallic mercury settled to the bottom of a body of water, posing no threat to the aquatic environment. Westoo (*13*), however, reported that 90 percent of the

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mercury in tissues of Swedish fish was present as methylmercury, an organic form highly toxic to wildlife and people. Johnels et al. (*14*) subsequently published the opinion that methylmercury could be created anaerobically by bacterial action in bottom sediments. This theory was verified by Jernelov and Jensen (*15*).

A 1971 study by the Geological Survey, U.S. Department of Interior, on surface water and sediments from the streams, canals, drains, and lakes in and below Brunswick Canyon, reported that substantial amounts of mercury from pre-1900 milling activity had entered the Carson River drainage system. Total mercury levels as high as 20.0 ppm were reported for bottom-sediment samples collected near the upstream end of Lahontan Reservoir; the highest level in sediment from the Carson River near Fort Churchill was 11.0 ppm. Attention immediately focused on the accumulation of mercury by domestic plants and animals raised in the area. A study was undertaken in 1971 by the College of Agriculture Extension Service, University of Nevada, Reno (*16*).

Equally important was the determination of mercury levels in tissues of native fish populations because aquatic organisms are known to concentrate mercury (*17-18*). Although numerous studies on mercury levels in fish of contaminated areas exist (*19-21*), data for the Carson River system were sparse and inconclusive (*22*). Previous work was concerned with data collected only from Lahontan Reservoir, a project completed in 1915 by the U.S. Department of Interior—Reclamation Service (predecessor of the Bureau of Reclamation). No references to fish size or weight had been published, nor had any attempt been made to correlate mercury levels in sediment with those in fish. Thus the present study was undertaken to ascertain total mercury levels for water and accompanying sediment, the relation between individual fish species and mercury levels, and the effect of fish weight on individual mercury concentrations in the Carson River drainage system.

The Carson River flows in a northeasterly direction from its origin in the Blue Lake area, Alpine County, Calif. (elevation 3,176 m). From the convergence of the east and west forks between Genoa and Minden, Nev., the river drops approximately 243 m to its termination at the Carson sink located 12.5 km north of Fallon, Nev. (elevation 1,173 m). Flow rates range from an average monthly high of 320 m^3/sec during maximum discharge in May, to an average monthly low of 4 m^3/sec in August (*23*). Bottom sediment ranges from coarse sand to clay, with turbidity and alkalinity generally increasing as the stream nears Lahontan Reservoir 10 km west of Fallon (*24*).

Initially, a 22-km stretch of the Carson River drainage system between New Empire, Nev., and Lahontan Reservoir was chosen for sampling. On the basis of presumed total mercury content of bottom sediment from pre-1900 ore milling, five collecting sites were designated representative of mercury content increasing progressively downstream. Areas were chosen for sampling on the basis of accessibility and previous data (*23*).

Sampling sites are shown in Figure 1. Site 1 was located 2.5 km east of Carson City, Nev., at the Brunswick Canyon Bridge. River width was approximately 24 m; the bed sloped to a maximum depth of 1.25 m. Water was moderately turbid and the bottom substrate was composed of materials ranging from very coarse sand to silt. Protective rock cover was generally poor and aquatic vegetation was sparse. This site represented an uncontaminated area above the 12 millsites associated with the drainage system, showing no evidence of previous milling activity.

Site 2 was located 5 km east of Carson City in Brunswick Canyon. It was immediately adjacent to the Eureka millsite, approximately 3 km below site 1. The watercourse at this station narrowed markedly to about 18 m as the river cut through the steep canyon. Depth increased from less than 1 m to a maximum of 2.12 m and bottom sediment was characterized by rock rubble. The current was considerably faster than it was upstream because of the river's decreased width and increased elevation decline. Jutting rock formations provided maximum shelter for fish and other aquatic organisms. Tailings from three Comstock milling operations extended to the water's edge. This area had been directly contaminated by pre-1900 milling activities.

Station 3 was located 1.25 km below Dayton, Nev., at the river's maximum width of 45.5 m. Depth varied from 1 to 2 m, channeling was prevalent, and turbidity generally increased as the waterflow slowed to a minimum for the four sampling sites. The bottom substrate was composed of sand ranging from coarse to very fine, with a brownish top coating of organic material. Although rock rubble was scarce, rooted vegetation was heavy and supplied adequate protective cover for aquatic organisms. Irrigation diversions for agriculture became numerous downstream from this site. No evidence of milling activity was observed near station 3 although a sand and gravel operation was located approximately 0.6 km upstream.

Site 4 was located 10 km east of Dayton midway between Break-A-Heart Ranch Number 1 and Break-A-Heart Ranch Number 2. The site was situated in the center of farmland and was characterized by numerous irrigation diversions. The river widened to approximately 24.2 m and reached a maximum depth of 2 m. The bottom substrate was mostly fine sand with little rock rubble. Banks were steep and channeled. No rooted vegetation was observed although flow was calm and turbidity moderate. This area represents a section of the river indirectly contaminated by milling operations.

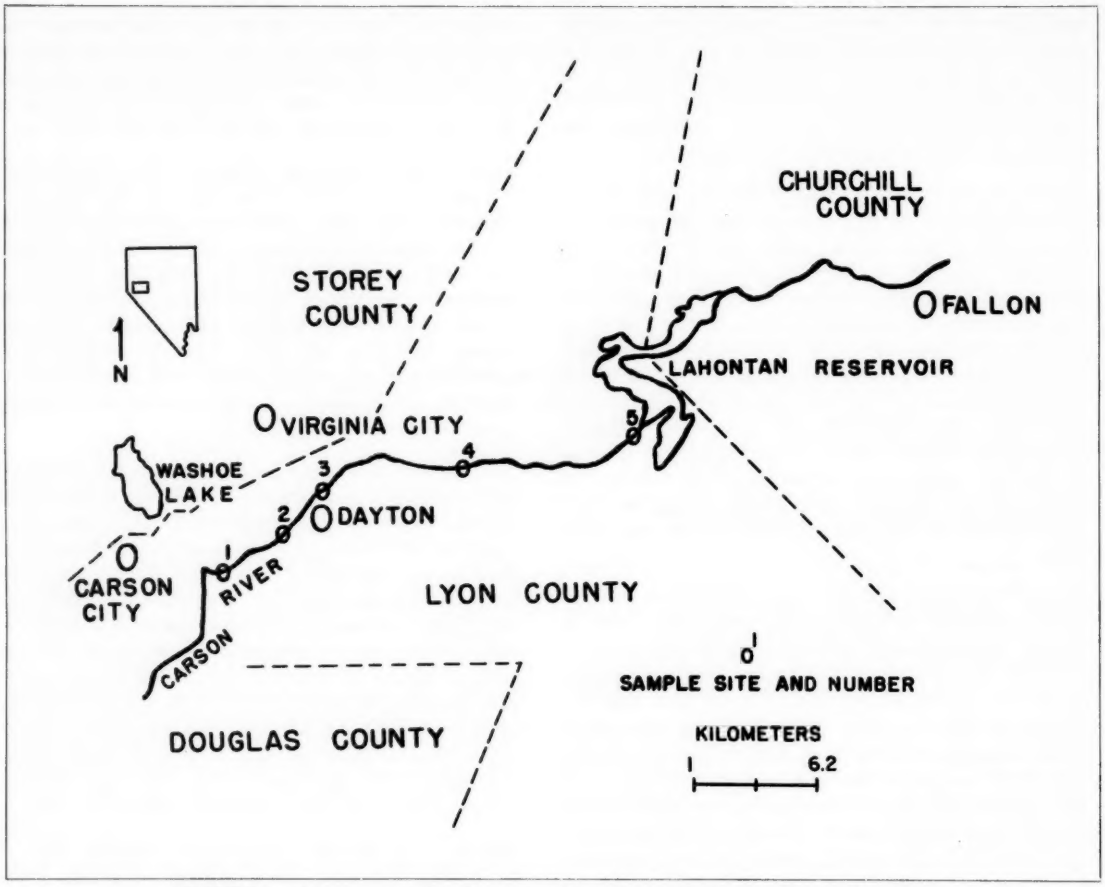

FIGURE 1. *Mercury sampling locations, Carson River, Lyon County, Nevada—1972*

Sampling site 5 was located at the upstream end of Lahontan Reservoir, which is located approximately 10 km west of Fallon, and is 10.5 km long. At conservation level during an average water year (under normal rainfall conditions), Lahontan impounds some 293,000 acre-feet of water, reaching depths in excess of 30 m (*24*). Bottom sediment is composed of clay, silt, and some fine organic material. This site represents a location remote from early milling operations.

Materials and Methods

SAMPLING

Samples of fish including crayfish were collected by electrofishing from the four Carson River sites during the months of July through October 1972, with a dual-electrode shocking apparatus. The electrical source was a 115-volt AC Briggs and Stratton gasoline-powered generator.

Each sampling effort consisted of a run between two predetermined points at the particular collecting site. Stunned fish were retrieved with a long-handled dip net. Emphasis was placed on collecting the largest fish of as many species as possible from each site.

The electrofishing gear was effective for the river collections but proved inadequate at the reservoir. Sampling at this site consisted of individual angler catch trolling and bankfishing; species numbers and size of fish varied uncontrollably.

Samples were labeled according to wet weight, species identification, and date and site of collection. They were preserved by freezing in water-filled polyethylene bags.

Bottom sediment samples collected in July 1972 from the four river sites consisted of composites of the uppermost 2.5-7.6 cm of fine-grained sediment from the right bank of the river. Samples for the Lahontan station were obtained in the shallows near the upstream end of the reservoir. All sediments were collected in acid-rinsed quart jars, characterized by sediment type, and labeled according to location. Unfiltered samples were preserved by acidification with 10 ml 1:1 distilled

water:nitric acid of low mercury content, and refrigerated until analysis (*25*).

Special attention was given to the stream flow rate during the sampling period because total mercury content of sediment and surface waters is related to stream discharge (*23*). Flow rates were supplied by the Geological Survey office in Carson City.

Surface water samples were also collected in July. Five dipped samples were taken across the stream from each site at a depth of approximately 15 cm, placed in acid-rinsed quart jars, dated, and preserved for future analysis by acidification with 10 ml 1:1 distilled water:nitric acid. Samples were then refrigerated. Water aliquots were not filtered because they were to be tested primarily for total mercury content.

Analytical Procedures

Cold vapor, flameless atomic absorption was selected as the analytical technique. Design samples were analyzed in triplicate with a Beckman Atomic Absorption System/Beckman model DB-G grating spectrophotometer, and reported as mean values. If sample variation between any duplicate set exceeded 5 percent, analyses for that set were repeated.

General procedures for fish samples followed those of Hatch and Ott (*26*), as modified by Uthe et al. (*27*) and Armstrong and Uthe (*28*). A 0.1-0.5-g sample of flesh taken from the trunk region below the dorsal fin and above the lateral line was dissolved in a 250-ml flask with 5 ml of a 4:1 solution of concentrated sulfuric acid : nitric acid and was oxidized with 15 ml 6 percent $KMnO_4$ and 2 ml 6 percent $K_2S_2O_8$ after cooling. Excess permanganate was reduced with a 30 percent H_2O_2 solution and the sample was made to 100 ml with distilled water. The sample was further reduced with 4 ml 10 percent $SnCl_2$ and aerated through the atomic absorption apparatus. Recovery studies of spiked fish samples averaged 92.7 percent with a sensitivity of 0.02 ppm. Standard deviations were 0.040 for samples analyzed at the 0.05 ppm spike level and ± 0.050 for tissues containing 0.5 ppm total mercury.

For sediment analysis, a 0.5-g sample was digested with 2 ml of the sulfuric acid : nitric acid solution and permitted to stand for 5 hours. After cooling, an oxidation-reduction procedure identical to that employed for fish tissues was followed. Recoveries for seeded sediment samples averaged 89.4 percent, with standard deviations of ± 0.041 at 0.05 ppm and ± 0.068 at 0.5 ppm.

A modified version of the Federal Water Quality Administration provisional method (*25*) was employed for water analysis. Samples consisted of unfiltered 50-ml aliquots digested for 5 hours with 5 ml of the sulfuric acid:nitric acid solution. After cooling, the portions were oxidized with 1 ml of a 6 percent $KMnO_4$ solution and 2 ml of a 6 percent $K_2S_2O_8$ solution. Samples were then reduced with a 10 percent hydroxylamine hydrochloride solution and 4 ml of a 10 percent $SnCl_2$. Recoveries averaged 94.1 percent with a sensitivity of ± 0.065 at the 0.2 ppb level and ± 0.040 at the 2.0 ppb level.

Results

Total mercury levels in surface waters of the drainage system are listed in Table 1. Values shown were not corrected for recovery. Based on triplicate analyses of unfiltered samples collected during a mean stream flow of 10.1 m^3/sec, results for the Brunswick Canyon Bridge, Eureka Mill, and Dayton Bridge collecting stations showed mean total mercury concentrations of less than 0.20 ppb, the minimum detectable level for water.

TABLE 1. ***Total mercury levels in surface water, Carson River, Nevada—July 18, 1972***

Site No.	Source	Total Hg, ppb
1	Carson River, Brunswick Canyon Bridge, 2.5 km east of Carson City	<0.20
2	Carson River, Eureka Millsite, 5 km east of Carson City	<0.20
3	Carson River, Dayton, 1.25 km below Dayton Bridge	<0.20
4	Carson River, 10 km east of Dayton, below Break-A-Heart Ranch No. 1	0.87
5	Lahontan Reservoir, Lyon County, at upstream shallows	2.10

NOTE: Concentrations based on average of triplicate analysis of unfiltered samples. If no absorbance was detected, Hg level was recorded as <0.20 ppb, minimum detectable level in water.

Levels in water from the two downstream sites increased as the river neared Lahontan Reservoir. At site 4 levels averaged 0.87 ppb. Site 5 levels suggested an accumulation of mercury by surface water: mean total mercury level based on triplicate analysis reached 2.10 ppb. The Food and Drug Administration (FDA), U.S. Department of Health, Education, and Welfare, considers 5.0 ppb to be the maximum level acceptable in drinking water (*8*). Although normal background levels for unpolluted surface waters vary widely and are affected by many parameters, the 2.10 ppb concentration at the Lahontan site was well above the 0.03 ppb level presumed to be the mean natural mercury content for uncontaminated waters (*8*).

Total mercury levels of sampled bottom sediment are shown in Table 2; they are not corrected for recovery. Based on triplicate analysis of unfiltered samples collected during a mean discharge flow of 10.1 m^3/sec on July 18, 1972, levels at the five stations also suggested that mercury content increased as water flowed downstream farther from previous milling activity. Mercury concentrations at Brunswick Canyon Bridge, site 1, ranged from 0.111 to 0.130 ppm with a mean level of 0.122 ppm. Corresponding levels of total mercury at the Eureka millsite, site 2, were as high as 0.447 ppm and averaged 0.349 ppm.

TABLE 2. *Total mercury in fine-grained sediment, Carson River and Lahontan Reservoir, Nevada—July 18, 1972*

Site No.[1]	Total Hg, ppm	Source	Description
1	0.122	Off right bank, 25 m below bridge	Dark brown silt and clay
2	0.349	Off right bank, 5 m below millsite	Coarse silt and sand
3	0.209	Off right bank, 0.6 km below Dayton Bridge	Dark brown clay
4	0.722	Off right bank, 100 m below Break-A-Heart Ranch No. 1	Silt and clay
5	1.345	10 m off bank below water surface	Dark brown silt and clay

NOTE: All samples represent the top 2.5-7.6 cm sediment. Concentrations based on average of triplicate analysis of unfiltered samples.
[1] See Table 1 for specific site locations.

An exception to the trend of mercury levels increasing in a downstream direction was observed at site 3. Here the mean level was only 0.209 ppm. Although stream flow decreased at this site, no apparent cause for this decline was observed.

Total mercury levels at site 4 were markedly higher than those observed at Dayton. Concentrations averaged 0.722 ppm for the three samples analyzed. Levels of mercury at Lahontan averaged 1.345 ppm. Such information implies that variations in mercury distribution in sediment along the river are caused by the washing action of local currents, which progressively enrich the mercury content of bottom sediments in a downstream direction.

Six species of fish and one species of crustacean were collected between July and October 1972 (Table 3) when mean stream discharge was 13.4 cm. Wet weight, average mercury concentration, standard deviation, and total mercury range were recorded for the species at each site in Tables 4-8. Residues were not corrected for recovery.

TABLE 3. *Aquatic organisms sampled for mercury content, Carson River drainage system—July-October 1972*

Species	Site No.[1]
Golden shiner *(Notemigonus crysoleucras)*	1-4
Crayfish *(Pacificastacus liniusculus)*	1-4
Tahoe sucker *(Catostomus tahoensis)*	1-4
Carp *(Cyprinus carpio)*	1-5
White bass *(Morone chrysops)*	5
White catfish *(Ictalurus catus)*	5
Brown bullhead *(Ictalurus nebulosus)*	5

[1] See Table 1 for specfic site locations.

As was the case for water and sediment samples, total mercury concentrations for individual aquatic species increased as the river approached Lahontan Reservoir. Table 4 shows mercury concentrations in organisms collected at the Brunswick Canyon Bridge. Mercury concentrations ranged from 0.020 to 0.520 ppm for the 24 samples collected; 12.5 percent exceeded 0.50 ppm. Highest levels were present in carp, which had an average level of 0.258 ppm total mercury. Levels in crayfish ranged from 0.100 to 0.520 ppm; two of the five samples had residues in excess of 0.50 ppm. Mean total mercury contents for the golden shiner and the sucker were notably low, averaging 0.183 and 0.189 ppm, respectively. Only three samples analyzed from this site contained less than 0.020 ppm total mercury.

A paired t-test showed no significant difference between mean total mercury concentrations. Wet-weight range and sample size of individual species collected at this site were lower than those of species from other stations.

Mean total mercury levels of organisms from the Eureka Millsite are presented in Table 5. Residues were consistently higher than at the Brunswick Bridge in all species except golden shiners. Likewise, mean wet weights of all species except the shiner were consistently higher. The greatest increase in mercury concentrations appeared in carp: five of eight specimens ranging from 1.02 to 411.20 g had wet weights exceeding 0.50 ppm. Levels in crayfish averaged 0.435 ppm total mercury with a range of 0.114 to 0.732 ppm. Suckers at this site demonstrated slightly higher levels of mercury than at Brunswick Bridge. Overall, 16.2 percent of the samples contained residues in excess of 0.50 ppm total mercury; 10.8 percent represented levels less than 0.02 ppm, the minimum detectable concentration for fish.

At the Eureka site mercury levels in carp were significantly greater than in golden shiners and suckers ($p < 0.01$). Differences between residues found in all other species comparisons were insignificant.

At the Dayton collecting station residues in shiners decreased appreciably (Table 6) and had a mean mercury concentration of 0.097 ppm, the lowest recorded mean level of any species at any site. Mercury content in crayfish and suckers also dropped. Concentrations averaged 0.105 and 0.103 ppm, respectively. Levels in carp, however, remained relatively high. Of eight carp specimens analyzed, levels in six exceeded 0.50 ppm.

A t-test comparison of levels present at the Dayton site demonstrated no significant difference in mercury levels accumulated by shiners, crayfish, and suckers. Mercury concentrations in carp, however, were significantly greater ($p < 0.01$), averaging five times more total mercury than the other three species.

Table 7 demonstrates corresponding mercury levels in fish tissue samples from site 4. Although one would have expected mercury concentrations to decrease somewhat proportionately with distance from milling sites, levels actually increased substantially. Of particular interest were concentrations found in crayfish. Ranging

from 0.534 to 0.969 ppm, mercury content in all six specimens exceeded the FDA 0.50 ppm tolerance level. Higher residues averaging 0.524 ppm were also found in sucker tissues. Although wet weights for this species were considerably greater than those for suckers at any of the previous sites, the wet-weight range for crayfish at this station was relatively small. Mercury levels in shiners were consistent with those at the other river sites, ranging up to 0.318 ppm. Concentrations for carp also remained rather constant although there was a high individual total mercury level of 1.360 ppm. Of the 37 samples collected at site 4, 45.9 percent contained total mercury concentrations in excess of 0.50 ppm. This represents the highest levels found in fish tissue at any of the four collection sites along the Carson River.

Mean total mercury levels in crayfish, suckers, and carp were significantly greater than those in shiners at this station ($p < 0.05$). Concentrations in crayfish were also significantly greater than those in suckers ($p < 0.05$). Residue variances between crayfish and carp did not differ significantly from those between suckers and carp.

Total mercury levels for aquatic species collected from Lahontan Reservoir are shown in Table 8. Wet weights of all species were greater than those of samples taken from the four river sites. Likewise, mercury content in sample tissues was higher. Highest concentrations were observed in white bass in which levels ranged from 0.501 to 2.720 ppm. Of eight specimens analyzed, all contained residues exceeding 0.50 ppm. White catfish had a mean total mercury content of 0.394 ppm. Levels in carp analyzed from this site were also high. One specimen contained 1.087 ppm total mercury. Every sample from the reservoir contained mercury residues above 0.020 ppm; 62.5 percent exceeded 0.50 ppm.

TABLE 4. *Total mercury residues in aquatic organisms, Brunswick Canyon Bridge, Carson River, Nevada (site 1)—1972*

Species	No. Fish in Sample	Mean Wet Weight, g (±S.D.)	Wet Weight Range, g	Mean Total Hg, ppm (±S.D.)	Total Hg, ppm	Positive Samples, % [1]	Samples (%) Over 0.50 ppm
Shiners	6	6.66(±3.33)	2.50-10.50	0.183(±0.157)	0.020-0.356	67.0	0.0
Crayfish	5	13.98(±4.09)	9.60-20.05	0.211(±0.176)	0.100-0.520	100.0	40.0
Suckers	7	50.07(±36.93)	7.22-105.33	0.189(±0.118)	0.020-0.333	85.7	0.0
Carp	6	73.58(±76.19)	8.29-182.30	0.258(±0.180)	0.069-0.503	100.0	16.6

[1] Samples considered positive if total mercury residue exceeded 0.020 ppm, minimum detectable level in fish.

TABLE 5. *Total mercury residues in aquatic organisms, Eureka millsite, Nevada (site 2)—1972*

Species	No. Fish in Sample	Mean Wet Weight, g (±S.D.)	Wet Weight Range, g	Mean Total Hg, ppm (±S.D.)	Total Hg, ppm	Positive Samples, % [1]	Samples (%) Over 0.50 ppm
Shiners	10	5.19(±3.14)	1.75-10.53	0.172(±0.128)	0.020-0.353	80.0	0.0
Crayfish	5	29.59(±13.92)	10.45-42.50	0.435(±0.278)	0.114-0.732	100.0	20.0
Suckers	14	77.88(±53.50)	5.10-178.40	0.222(±0.104)	0.122-0.476	100.0	0.0
Carp	8	158.58(±207.21)	1.02-411.20	0.637(±0.367)	0.020-0.919	75.0	62.5

[1] Samples considered positive if total mercury residue exceeded 0.020 ppm, minimum detectable level in fish.

TABLE 6. *Total mercury residues in aquatic organisms, Dayton, Nevada (site 3)—1972*

Species	No. Fish in Sample	Mean Wet Weight, g (±S.D.)	Wet Weight Range, g	Mean Total Hg, ppm (±S.D.)	Total Hg, ppm	Positive Samples, % [1]	Samples (%) Over 0.50 ppm
Shiners	10	3.67(±1.25)	1.98-5.20	0.009(±0.069)	0.020-0.170	50.0	0.0
Crayfish	7	15.90(±5.66)	9.55-21.35	0.105(±0.054)	0.020-0.149	85.7	0.0
Suckers	14	42.97(±31.46)	14.95-105.10	0.103(±0.070)	0.033-0.250	100.0	0.0
Carp	8	109.36(±139.48)	1.59-400.20	0.536(±0.113)	0.355-0.650	100.0	75.0

[1] Samples considered positive if total mercury residue exceeded 0.020 ppm, minimum detectable level in fish.

TABLE 7. *Total mercury residues in aquatic organisms, Break-A-Heart Ranch No. 1, Nevada (site 4)—1972*

Species	No. Fish in Sample	Mean Wet Weight, g (±S.D.)	Wet Weight Range, g	Mean Total Hg, ppm (±S.D.)	Total Hg, ppm	Positive Samples, % [1]	Samples (%) Over 0.50 ppm
Shiners	11	2.43(±1.75)	0.87-6.31	0.136(±0.127)	0.020-0.318	54.5	0.0
Crayfish	6	13.24(±3.99)	9.20-20.20	0.756(±0.178)	0.534-0.969	100.0	100.0
Suckers	11	127.29(±69.22)	24.20-210.70	0.524(±0.227)	0.205-0.965	100.0	63.6
Carp	9	95.41(±167.11)	1.40-399.12	0.616(±0.385)	0.020-1.360	88.9	55.5

[1] Samples considered positive if total mercury residue exceeded 0.020 ppm, minimum detectable level in fish.

These data suggest that the elevated mercury levels in fish species sampled from Lahontan are probably due to the presence of fluvial sediment high in mercury content deposited at the upper end of the reservoir.

A statistical comparison showed that mean total mercury concentrations in white bass collected from Lahontan Reservoir were significantly greater than those in white catfish or brown bullhead ($p < 0.01$). No other comparisons of the various species demonstrated major differences in mercury levels although a larger sample size would be desirable for this observation to be considered significant.

A t-test was performed to evaluate variation of mean total mercury contents among species of the four Carson River sites. Results demonstrated several distinct trends. In general, total mercury concentrations in shiners did not vary significantly from site to site. Levels in crayfish collected at site 4, however, were significantly greater than those sampled at the other river sites ($p < 0.01$). Levels in suckers from this area were also significantly greater than those from the other river sites ($p < 0.01$). Residues in the Brunswick Bridge carp samples were significantly less than those in carp collected from downstream sites ($p < 0.05$).

A relationship also appeared between mercury concentrations and species' weight. Further statistical analyses were performed to determine whether there was a positive correlation strong enough to establish weight limits within which safe mercury concentrations could be found. Table 9 presents simple correlation coefficients for mercury versus weight. The relationship was examined for the seven species collected in the study area, although sample sizes for white bass, white catfish, and brown bullhead were too small for accurate prediction.

Several points are noteworthy. Among species, the relation between mercury concentrations and fish weight was not consistent. Within species, there was generally a strong positive correlation between mercury content and fish weight, as indicated by shiner, sucker, carp, brown bullhead, and white bass populations. The absence of such a correlation in white catfish is probably attributable to small sample size and wet-weight range. No such explanation was apparent for crayfish.

Statistical analyses were also performed to determine the relationship between mercury levels in bottom sediments and mean residues in individual fish species at each site (Fig. 2). Data demonstrated no significant correlation between mercury concentrations in shiners and carp and levels of accompanying sediments for the four river sites. There was, however, a significant correlation between mean residue levels in suckers and crayfish and mercury concentrations in bottom sediment at individual collecting stations ($p < 0.10$). Similar analyses for white bass, white catfish, and brown bullhead were not initiated because these species were collected only from Lahontan Reservoir. More individual sampling sites are necessary to insure validity of correlations between mean mercury concentrations in aquatic species and those in respective bottom sediments.

TABLE 8. *Total mercury residues in aquatic organisms, Lahontan Reservoir, Nevada (site 5)—1972*

Species	No. Fish in Sample	Mean Wet Weight, g (±S.D.)	Wet Weight Range, g	Mean Total Hg, ppm (±S.D.)	Total Hg, ppm	Positive Samples, % [1]	Samples (%) Over 0.50 ppm
White bass	8	414.23(±72.79)	320.80-527.20	1.297(±0.845)	0.501-2.720	100.0	100.0
White catfish	5	356.29(±76.73)	280.35-440.11	0.374(±0.227)	0.211-0.769	100.0	20.0
Brown bullhead	6	419.49(±112.54)	300.98-540.20	0.554(±0.394)	0.250-1.083	100.0	33.3
Carp	5	284.17(±126.97)	120.00-392.35	0.743(±0.285)	0.382-1.087	100.0	80.0

[1] Samples considered positive if total mercury residue exceeded 0.020 ppm, minimum detectable level in fish.

TABLE 9. *Analyses for homogeneity of mercury-weight conditions in aquatic organisms, Carson River drainage system, Nevada—1972*

Species	No. Fish in Sample	Mean Hg Content, ppm	Size, g	Correl. Coeff.	p-Value
Shiners	36	0.129	0.87-10.53	0.6818	0.001
Crayfish	23	0.345	9.20-42.50	0.0204	NS
Suckers	45	0.245	5.10-210.70	0.7633	0.001
Carp	36	0.547	1.02-411.20	0.6165	0.001
White bass	8	1.297	320.80-527.20	0.9750	0.001
White catfish	5	0.374	280.35-440.11	0.7746	NS
Brown bullhead	6	0.554	300.98-540.20	0.8971	0.02

NOTE: NS = not significant.

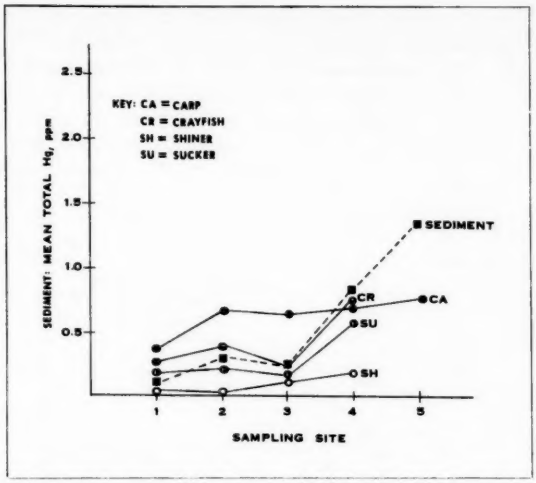

FIGURE 2. *Total mercury in fine-grained bottom sediment and aquatic organisms, Carson River, Nevada—1972*

Discussion

Mercury concentrations in the environment are difficult to assess. Before one declares a water body or organisms present in that water contaminated with mercury waste from people's activities, it is necessary to know the naturally occurring background levels of the metal in both water and organisms. Because the full extent of the mercury problem has only recently become apparent, attempts to establish background levels have been limited. Background data on mercury concentrations in the Carson River consist of surface water and sediment analyses taken upstream from pre-1900 ore milling sites in 1971-72 by the Geological Survey. No previous information exists on baseline levels present in Carson River fish, although random spot samplings of various locations throughout the State were performed by the U.S. Environmental Protection Agency in cooperation with the Nevada State Fish and Game Commission in 1970-71. In general, mercury content for surface water above the Brunswick Canyon area is less than 0.20 ppb (*23*). Total mercury present in corresponding sediments ranges between 0.04 and 0.10 ppm. These concentrations compare with the accepted 0.03 ppb mean natural background level for uncontaminated stream and river waters and the 0.073 ppm mercury content found in uncontaminated stream and river sediments (*8*).

The present study and the Geological Survey findings (*23*) indicate that mercury concentrations in the Carson River drainage system downstream from pre-1900 gold and silver mines far exceed naturally occurring background levels in the river. Apparently total mercury in water and sediment increases in proportion with increased distance downstream from early milling sites. This trend is presumably due to contributions from milling tailings along the river which progressively enrich mercury content downstream, reflecting the flushing action of seasonal runoff. Greatest concentrations are within and immediately upstream from Lahontan Reservoir.

Mercury concentrations in water and sediment are greatly influenced by stream flow because an increase in flow enhances scrubbing action on geological deposits. Samples collected during periods of high discharge generally reflect higher mercury content (*23*). This is evidenced by contrasting values of water and sediment collected from the Carson River by the Geological Survey during a period of high snow runoff in May with those recorded in this study from similar sites during the months of minimum discharge, July and August 1972. Of six surface waters sampled by the Geological Survey, total mercury content ranged from 0.2 to 6.3 ppb. Two samples exceeded the FDA 5.0 ppb tolerance limit for drinking water. Corresponding sediment analyses revealed mercury levels ranging from 9.5 to 20.0 ppm. Concentration in samples from the present study showed total mercury content ranging from 0.20 to 2.10 ppb for water, and from 0.122 to 1.345 ppm for sediment. Extrapolation of these data suggests that mercury content for surface water and sediment can vary markedly, depending on stream discharge. On this basis, it can further be assumed that since maximum discharges during 1971-72 were well below those of many recent years, peak mercury concentrations during the same years were probably also less than those associated with higher flows (*23*).

The ability of aquatic organisms to concentrate mercury above the levels found in their environment is well known (*17,29*). Mechanisms by which fish accumulate organomercury compounds, however, are not fully understood. It is believed that all microorganisms capable of vitamin B^{12} systhesis are also capable of methyl and dimethylmercury synthesis (*30*). Apparently carbon, phosphates, nitrogen, and various trace elements provide these organisms with the food they require to grow and multiply. This food supply in turn determines the size of populations of bacteria and molds present, and thus the rate of organic mercury conversion. The reaction is viewed as a detoxification of the microorganisms' environment at the expense of the fish, and is important because the solubilities of these organomercurials permits the compounds to be directly absorbed in fish by diffusion across the gills (*31*).

Numerous investigators have shown mercury residue in fish tissues to be present as methylmercury. Kamps et al. (*32*) demonstrated that methylmercury comprises 95 percent of the total mercury present in white bass. Westoo has shown mercury in pike to be at least 90 percent methylmercury (*13*). Although a detailed evaluation by the Geological Survey showed most mercury in the water and bottom sediments of the Carson River present as a component of the sulfide or nonmethyl

organic substance (*23*), it became apparent after analyzing some 200 fish samples in the present study that methylmercury conversion was occurring in the system. Results in Table 8 demonstrate mercury concentrations in white bass and brown bullhead collected from Lahontan Reservoir exceeding 2.70 and 1.08 ppm, respectively. Levels for carp and crayfish from the Carson River near site 4 ranged from 0.20 to 1.360 ppm, averaging 0.616 and 0.756 ppm, respectively. These data represent a substantial uptake of mercury from the aquatic environment, the majority of which must be assumed methylmercury. These levels also represent residues which are considerably higher than 0.20 ppm, generally accepted as the naturally occurring background level for most fish (*8*).

As noted earlier, residues varied widely between individual fish within a species. This was particularly evident in carp from site 4, whose individual mercury residues ranged from 0.020 to 1.360 ppm, and in white bass from Lahontan, whose levels varied from 0.501 to 2.720 ppm. This variance appeared to be, in part, a function of fish weight as demonstrated by the highly significant correlations between fish weight and total mercury content shown in Table 9. Similar studies have also shown significant correlations between fork length and total mercury content, and age and mercury accumulation for several species of fish (*32,33*). Neither of these relationships was explored in the present study.

In order to predict weight ranges with a mercury content less than 0.50 ppm, regression analyses were performed on species which reflected highly significant correlation between mercury content and weight. Values for intro-species regressions of total mercury appear in Table 10; estimated regression lines for each of the five species are shown in Figure 3. Extrapolation of these data permits prediction of individual mercury levels within species, providing wet-weight values are given.

For example, it can be predicted that the total mercury content of a carp weighing 200.0 g corresponds to 0.623 ppm. This prediction is reached by direct interpretation from the carp regression line (Fig. 3); or by calculation from the equation $Y = a + b(X)$, substituting the values 0.395 for a, and 1.139×10^{-3} for b (Table 10), and 200.0 g for X. Similar computations show the following practical wet-weight limits below which individuals of the five species can be expected to contain less than 0.500 ppm total mercury: shiner, 18.00 g; sucker, 180.00 g; carp, 100.00 g; brown bullhead, 430.00 g; and white bass, 350.00 g.

Sample sizes for white bass and brown bullhead were too small for accurate prediction. A wider range would also have been desirable because predicted values are likely to fall far outside the sample range. The merit of such predictions might possibly be found by selective fishing; the person fishing could weigh each fish to determine mercury content.

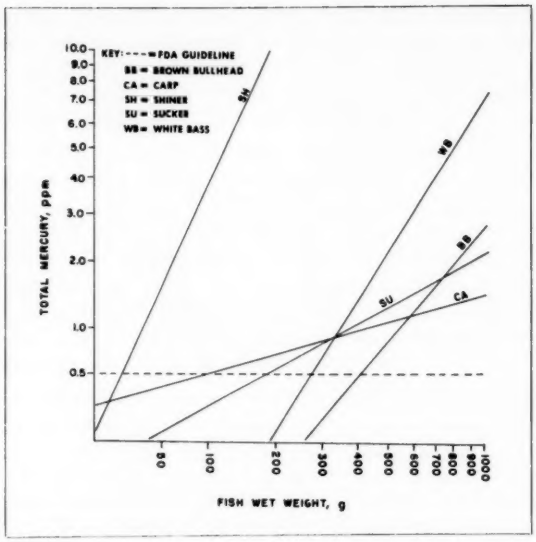

FIGURE 3. *Linear regressions of total mercury content on fish wet weight, Carson River, Nevada—1972*

TABLE 10. *Regression equations for total mercury content versus wet weight in fish, Carson River drainage system, Nevada—1972*

Species	No. Fish in Sample	Total Mercury Content [1]
Shiner	36	$7.623 \times 10^{-3} + 2.955 \times 10^{-2}$(FW)
Sucker	45	$6.290 \times 10^{-3} + 2.416 \times 10^{-3}$(FW)
Carp	36	$0.395 + 1.139 \times 10^{-3}$(FW)
White bass	8	$-3.284 + 1.115 \times 10^{-2}$(FW)
Brown bullhead	6	$-1.121 + 4.016 \times 10^{-3}$(FW)

[1] FW = fish weight, g.

Another conclusion was that mercury levels varied widely between species, indicating marked differences in ability to concentrate mercury (Fig. 4). There is also increasing variance between species and increasing frequency of mean levels exceeding 0.50 ppm total mercury in a downstream direction. These differences are probably explained by variations in feeding activities of the different groups and the accompanying mercury content in surface water and bottom sediments of the respective stations. Levels in golden shiners ranging from 0.097 to 0.183 ppm were consistently lower than concentrations found in other species at each site. McKechnie has shown that algae and plankton are the principal food of shiners when available (*34*). Algae and plankton represent minimum mercury sources because of their position at the base of the food pyramid.

Mean total mercury levels for suckers and crayfish were also consistently low from site to site. In general, mean mercury concentrations in suckers ranged from 0.103 to 0.524 ppm and those for crayfish ranged from 0.105

to 0.756 ppm. The exception for both species was observed at site 4, where t-test comparisons showed mean mercury residues of 0.524 ppm for suckers and 0.756 ppm for crayfish. These were significantly greater than levels in the other species from the site ($p < 0.01$) and mean values for individuals of the same species at the three upstream sites ($p < 0.01$).

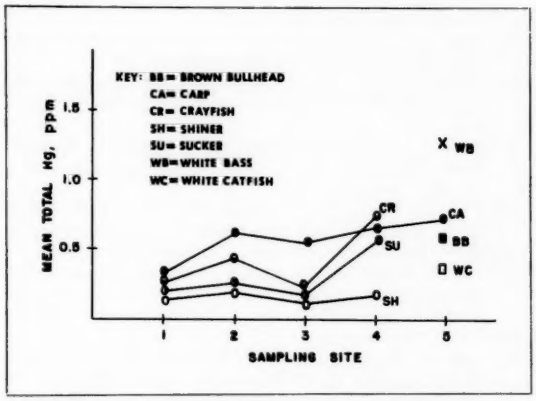

FIGURE 4. *Mean total mercury in aquatic organisms, Carson River, Nevada—1972*

This again reflects feeding activities. Suckers are bottom-dwellers, grazing mainly on green algae, insects, and mollusks (*35*). Crayfish are also bottom-dwellers, feeding on nearly anything that is edible (*24*). Because total mercury content of surface water and sediments was also significantly higher at site 4, and because fish have been shown to absorb mercury compounds directly through their gills and through ingestion of food (*31*), authors had expected tissue levels of bottom-feeders to be higher as well.

Average residues in carp ranged from 0.258 to 0.743 ppm in the five collecting stations. A t-test comparison demonstrated significantly lower levels ($p < 0.01$) in carp taken from the New Empire location above pre-1900 milling activity. Further t-test comparison showed mean mercury concentration in carp significantly higher than in shiners except at site 1 ($p < 0.01$). Mean concentrations varied insignificantly from mean content of other species at individual stations.

Authors made these comparisons with the hope of arriving at a single fish species as an indicator of trends in mercury levels within a station or the entire drainage system over a period of years. Carp, then, represent the species most suitable for such an indicator in the Carson River system: they are present at all five sampling sites; mercury levels within the species do not vary significantly except above milling operations where evidence of natural background concentration exists; and mercury concentrations in carp are similar to those in the other species except shiners.

Carp also reflect an increasing omnivorous diet. Filamentous green algae make up the bulk of their food (*36*); aquatic insects, crustaceans, and small fry are secondary items. Some large carp are even cannibalistic, as demonstrated in downstream sites where most of the large carp were captured by angling with live minnow bait. The species, therefore, provides a good index of mercury accumulation for fish that are both herbivorous and omnivorous.

Because mercury levels in white catfish, brown bullhead, and white bass were determined only for samples collected from Lahontan Reservoir, a comparison of mercury variation among sites was not possible. A t-test comparison of the three species sampled at site 5 showed that mean levels in white bass were significantly higher than in brown bullhead and white catfish ($p < 0.05$). Concentrations of 0.554 ppm in brown bullhead and 0.374 ppm in white catfish varied insignificantly. The differences between mercury levels in the three species can again be explained in terms of feeding habits. White catfish and brown bullhead are omnivorous, feeding near the bottom. Major food items include algae, diatoms, and crustacea; secondary items include insects and other fish (*24*). White bass are primarily piscivorous, consuming a greater volume of fish than all other foods combined (*37*); this habit was reflected in their average total mercury level of 1.297 ppm. Accompanying sediment and water analyses from the reservoir were correspondingly high, undoubtedly influencing mercury uptake by fish.

D'Itri has categorized fish according to their ability to accumulate mercury (*8*). Category I, encompassing predators such as bass and pike, accumulate the highest quantities of mercury. Category II includes such fish as carp and bluegills, which usually contain less mercury. Fish in category III, generally including bottom-feeders such as catfish and suckers, have the least tendency to concentrate mercury. This categorization is corroborated by observations in the present study, especially from Lahontan Reservoir, where mean total mercury content for white bass averaged 1.297 ppm. Category II carp demonstrated a mean content of 0.743 ppm at site 5 although one specimen at this site contained 1.087 ppm. Category III catfish, brown bullhead, averaged 0.554 ppm total mercury at site 5. Results imply biological magnification of mercury among fish or an increase in levels of mercury in fish food organisms at each trophic level of the generalized food chain.

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ERRATA

PESTICIDES MONITORING JOURNAL, Volume 8, Number 4, pp. 247-254. In the paper "Organochlorine Residues in Starlings, 1972," the following corrections are in order:

Page 250

Column 2, line 6, should read, "Residue values of DDT and its metabolites from each station were compared, as were those of dieldrin, for the periods summer-winter 1967-68 versus fall 1968, fall 1968 versus fall 1970, and fall 1970 versus fall 1972."

Table 2, title, should read, "Geometric and arithmetic means of DDT and dieldrin residues in starlings, 1967-72."

Page 251

Column 2, line 1: "periods" should be "period." Column 2, lines 16-17: "metabolic" should be "metabolite."

Page 252

Table 5: footnotes 1 and 2 should be reversed; the former refers to sites and the latter to PCB's.

Page 254

Column 2, line 1, should read: "A metabolite of chlordane, oxychlordane, was found in nearly all samples at very low levels."

Column 2, Literature Cited, reference 1: publication date should be 1969.

APPENDIX

Chemical Names of Compounds Discussed in This Issue

ALDRIN	Not less than 95% of 1,2,3,4,10,10-Hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-*endo-exo*-5,8-dimethanonaphthalene
BHC (BENZENE HEXACHLORIDE)	1,2,3,4,5,6-Hexachlorocyclohexane (mixture of isomers). Commercial product contains several isomers of which *gamma* is most active as an insecticide.
CHLORDANE	1,2,3,5,6,7,8,8-Octachloro-2,3,3a,4,7,7a-hexahydro-4,7-methanoindene. The technical product is a mixture of several compounds including heptachlor, chlordene, and two isomeric forms of chlordane.
DDD	See TDE.
DDE	Dichlorodiphenyl dichloro-ethylene (degradation product of DDT) *p,p'*-DDE: 1,1-Dichloro-2,2-bis(*p*-chlorophenyl) ethylene *o,p'*-DDE: 1,1-Dichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethylene
DDMU	1,1'-(Chloroethenylidene)bis(4-chlorobenzene)
DDT	Main component (*p,p'*-DDT): α-Bis(*p*-chlorophenyl)β,β,β-trichloroethane Other isomers are possible and some are present in the commercial product. *o,p'*-DDT: [1,1,1-Trichloro-2-(*o*-chlorophenyl)-2-(*p*-chlorophenyl) ethane]
DIELDRIN	Not less than 85% of 1,2,3,4,10,10-Hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-*endo-exo*-5,8-dimethanonaphthalene
ENDOSULFAN	6,7,8,9,10,10-Hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiepin 3-oxide
ENDRIN	1,2,3,4,10,10-Hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-*endo-endo*-5,8-dimethanonaphthalene
HCB	Hexachlorobenzene
HEPTACHLOR	1,4,5,6,7,8,8-Heptachloro-3a,4,7,7a-tetrahydro-4,7-*endo*-methanoindene
HEPTACHLOR EPOXIDE	1,4,5,6,7,8,8-Heptachloro 2,3-epoxy-3a,4,7,7a-tetrahydro-4,7-methanoindane
METHOXYCHLOR	1,1,1-Trichloro-2,2-bis(*p*-methoxyphenyl) ethane
MIREX	Dodecachlorooctahydro-1,3,4-metheno-2H-cyclobuta[cd]pentalene
NONACHLOR	1,2,3,4,5,6,7,8-Nonachlor-3a,4,7,7a-tetrahydro-4,7-methanoindan
OXYCHLORDANE	2,3,4,5,6,6a,7,7-Octachloro-1a,1b,5,5a,6,6a-hexahydro-2,5-methano-2H-indeno(1,2-β)oxirene
PCB'S (POLYCHLORINATED BIPHENYLS)	Mixtures of chlorinated biphenyl compounds having various percentages of chlorine
TDE	2,2-Bis(*p*-chlorophenyl)-1,1-dichloroethane
THIODAN	See endosulfan.
TOXAPHENE	Chlorinated camphene (67-69% chlorine); product is a mixture of polychlor bicyclic terpenes with chlorinated camphenes predominating.

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